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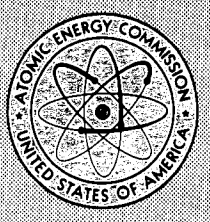
RADIOACTIVE FALLOUT FROM NUCLEAR WEAPONS TESTS

Proceedings of a Conference Held in Germantown, Maryland

November 15-17, 1961

BOOK 1



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BIOLOGY AND MEDICINE

RADIOACTIVE FALLOUT FROM NUCLEAR WEAPONS TESTS

PROCEEDINGS OF A CONFERENCE

NOVEMBER 15-17, 1961

Held in the

U. S. Atomic Energy Commission Auditorium

Germantown, Maryland

Under the auspices of
Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission
Washington 25, D. C.

Edited by

Alfred W. Klement, Jr.

February 1962

ABSTRACT

Technical papers are presented which review the various research projects, sponsored by the AEC, related to fallout from weapons tests. Reviews of specific related programs by representatives from Canada and the United Kingdom are also included. The scope of the conference includes: (1) characteristics of fallout, (2) atmosphereic influences on deposition of local and global fallout, (3) distribution and cycling of radionuclides in the environment, and (4) fallout nuclides in the food chain and man. Discussion of each topic by authors and other participants is included.

FOREWORD

From time to time conferences and symposia are held to review and discuss topics of major interest to the Commission. This conference on radioactive fallout deals with one of the important areas of research supported by the Division of Biology and Medicine. It is hoped that this conference and the report of its proceedings will be informative and useful to the participants as well as to other interested groups not in attendance. It is hoped also that these proceedings together with those of other conferences related to the Division's program will assist in the overall evaluation of fallout problems and will point out areas requiring further research.

I wish to express my appreciation to the speakers and to the other participants for giving of their time in order to make this conference possible.

Charles L. Dunham, M.D., Director
Division of Biology and Medicine

CONFERENCE
ON
RADIOACTIVE FALLOUT FROM NUCLEAR WEAPONS TESTS

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*Denotes speakers for multiple-author papers

Acknowledgements

We wish to acknowledge with thanks the assistance of several individuals, who, together with the participants, helped make a successful conference possible. Some of these include: Division of Headquarters Services for auditorium arrangements, and especially Mr. Paul Barrett, the projectionist, Mr. Donald P. Huddleston for operation of the sound system, and Mr. Robert J. Wright for transportation arrangements; Mr. Robert Brothers and members of the Administrative Branch of the Division of Biology and Medicine for assisting with many of the preparations; Mrs. Geogetta Blasingame, Mrs. Leona von Bretzel, and Mrs. Harriett Mohler for their assistance in the preparation and conduct of the conference and in the publication of the proceedings.

INTRODUCTORY REMARKS

Joshua Z. Holland, Chief
Fallout Studies Branch
Division of Biology and Medicine
U.S. Atomic Energy Commission
Washington, D. C.

This conference continues a series of conferences of active workers in the U.S. AEC fallout research program* and its close associates. The last one was held at the AEC Health and Safety Laboratory in New York in September 1960 and was a relatively informal meeting with no proceedings issued. Earlier conferences were closely associated with the 1959 Fallout Hearings of the Joint Committee on Atomic Energy, the pre-and post-HARDTACK planning of sample collection and analysis, and, still earlier, the periodic "Sunshine meetings" called by Dr. Libby.

When the planning of this conference was begun over 6 months ago we did not expect that fallout would be a "hot" subject. It was felt that this would be a good time to call together the people who had been working in the different laboratories, to take stock of the new knowledge and understanding that had been gained and to consider the specific questions which remain unanswered. We did plan a more formal and more comprehensive program than usual. Several of the major projects, particularly those involving nation-wide or global surveys of radioactivity in soils, diets, and human bone were coming to an end in the particular form in which they had been carried on up to that time, and considerable reorientation of the program was anticipated. The two main purposes of the conference were and still are to promote technical discussion on an unclassified basis among the scientists working on various parts of the fallout problem, and to produce in the proceedings an organized compendium of the new information gained on this subject since the 1959 Fallout Hearings.

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* This program, including current research projects, is described in detail in U.S. AEC report TID-12616, Atmospheric Radioactivity and Fallout Research, April 1961. 72 pp. (Available from the Office of Technical Services, U.S. Dept. of Commerce, Washington 25, D.C. $0.75).

The subject matter of the conference includes the history of nuclear weapons debris and its nuclear radiations from the time of detonation to the time when people are exposed to the radiation in any way. Radiation effects are not included nor are the basic biological mechanisms of metabolism and transport of radionuclides. Survey results are not to be presented as such but only in so far as they are used to support scientific findings.

Much of the work to be reported on has been supported under the program of the Fallout Studies Branch. Summaries and reviews of work supported by other Branches of the Division of Biology and Medicine as well as some work conducted under the auspices of other AEC divisions are also included for completeness of coverage. Visitors from the United Kingdom and Canada have also been invited to present summaries of research results. However, of the United States agencies conducting studies of fallout, only the program supported by the AEC is being reviewed in this symposium.

In short, we feel that in addition to the scattered journal articles and monographs in which the material to be presented at this symposium would eventually appear, we have an obligation to collect and make available the fruits of our research program on this important subject in more concise summary form. We could not think of a better way to do this than to ask each of the principal research scientists to summarize his own area of work. Review and discussion would clearly add to the depth and breadth of treatment and particularly to the identification of unsolved problems. And so, our present conference.

PHYSICAL, CHEMICAL, AND RADIOLOGICAL CHARACTERISTICS OF FALLOUT

Session Chairman: Harold A. Knapp
Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission

NEVADA TEST SITE FALLOUT: SOME CHARACTERISTICS, ITS APPARENT ENVIRONMENTAL EQUILIBRIUM AND BIOLOGICAL AVAILABILITY

Kermit H. Larson, Howard A. Hawthorne, and John H. Olafson
Laboratory of Nuclear Medicine and Radiation Biology
Department of Biophysics and Nuclear Medicine
University of California at Los Angeles
Los Angeles, California

Fallout from the detonation of test devices at the Nevada Test Site is governed by many complex variables such as: energy yield and type of device, wind structure, support used for the device, nature of the ground surface, how much contact the fireball has with the ground surface, and mass of inert material surrounding the device. Some of our findings related to the distribution and characteristics of fallout from Nevada Test Site operations are summarized in the following statements. We will restrict this summary to fallout from detonations of tower mounted devices and from balloon mounted or air dropped devices all having less than 75 KT yield.

Some Characteristics of NTS Fallout Patterns

The coordination of aerial radiation survey measurements of fallout patterns with ground survey meter measurements increased the detail and accuracy of fallout pattern delineation as well as the distances to which fallout patterns could be documented from the point of detonation.

By use of aerial radiation survey equipment and techniques as developed by the U. S. Geological Survey and Davis and Reinhart from the Oak Ridge National Laboratory, fallout radiation intensities within an area of about 10,000 $mi.^2$ were measured in less than 12 hours by one aircraft. When these data are properly reduced aerial measurements agree within $\pm$ 10% of measurements taken 3 ft. above the ground by conventional survey meters.

These studies were supported by Contract AT(04-1)GEN-12 between the Atomic Energy Commission and the University of California.

During Operation PLUMBBOB (1957), we routinely measured fallout patterns to distances of 200 to 300 mi. from Ground Zero; however, one fallout pattern from a tower-supported detonation was documented as far as 700 mi. from the Nevada Test Site with the radiation levels readily detectable at that distance.(1)

The detailed documentation of fallout patterns during Operation PLUMBBOB (1957) afforded the opportunity to confirm the existence of "hot spots" in most fallout patterns. "Hot Spots" were first identified in 1948 when the fallout pattern of the Operation TRINITY detonation in New Mexico had been outlined in detail.(2)

While the occurrence of "hot spots" has been associated with prominent terrain features in many cases, sufficient data are not available to explain their mechanism of formation or to permit their prediction.

Fallout from aerial bursts has not been detectable by conventional ground survey methods(3) and is difficult to document even with aerial survey methods within 200 mi. of Ground Zero(1). Two test devices detonated from balloons at 1500 ft. without the fireball intersecting the ground surface deposited less than 0.2% of the theoretical fallout radioactivity.* The area measured is defined in this case by the 0.1 mr/hr radiation intensity contour (at H + 12 hrs.) between the distance of 1 mi. from Ground Zero and this distance corresponding to a fallout time of H + 12 hrs. To illustrate the effect of intersection of fireball, a balloon-supported detonation in which the fireball only touched the ground surface deposited 2.12% of the theoretical fallout within the 1 mr/hr contour to H + 12 hr. fallout time. However, fallout originating from test devices mounted on steel towers whose fireballs in some cases intersected the ground surface to perhaps 1/4 of their apparent diameter, and in other cases only touched, deposited 6.7 to 24.5% of the theoretical fallout radioactivity within the same area limits. These data are presented in Table 1.

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*The theoretical fallout is calculated on the basis of 300 gamma Mc at H + 1 hr. per KT yield.

TABLE 1

Per cent deposition of total fallout, Sr^{89} and Sr^{90} within limits of 1 mi. from Ground Zero to H + 12 hr. fallout time, Operation PLUMBBOB (1957)

Shot	Yield (KT*)	H + 12 hr. Distance (mi.)	Per cent deposited Total Fallout	Sr89	Sr^{90}
Tower Mounted Shots					
Fizeau	11.1	160	6.67	0.45	1.59
Galileo	11.4	83	10.4	0.80	2.82
Boltzmann	11.5	213	24.5	1.80	6.33
Shasta	16.5	151	21.7	2.052	7.18
Diablo	17.0	146	16.6	1.22	4.27
Whitney	18.5	87	16.1	1.61	5.65
Smoky	44.0	238	19.6	1.70	5.97
Balloon Mounted Shots					
Wilson	10.3	92	0.91	0.04	---
Newton	12.0	200	0.10	0.004	---
Priscilla	36.6	260	2.12	0.13	---
Hood	74.3	366	0.17	0.008	---

*Reference (4)

Deposition of Radiostrontium in Areas Adjacent to NTS

Table 1 also gives the various estimated percentages of Sr^{89} and Sr^{90} deposited within the defined area of fallout for several shots of Operation PLUMBBOB(1). Approximately 0.13% of the total amount of Sr^{89} produced by a balloon-supported detonation whose fireball just intersected the ground surface was deposited within the fallout time of arrival of H + 12 hrs. On the other hand, only 0.004 and 0.008% of the total Sr^{89} produced was deposited within H + 12 hrs. fallout time by two balloon-supported detonations whose fireballs did not intersect the ground. Tower-supported detonations deposited from 0.5 to 2% of the Sr^{89} produced and from 1.6 to 7.2% of the total Sr^{90} produced within H + 12 hrs. fallout time.

These calculations were based on the results of analyses of fallout debris samples for Sr^{89} and Sr^{90} and integrated fallout radiation intensities converted to curies by ratios of $\mu c/ft.^2$ and mr/hr.

The percentage deposition of Sr^{89} in fallout from tower-mounted devices was less than that of Sr^{90} out to distances corresponding to H + 12 hr. fallout arrival time. This is attributed to relatively low percentages of Sr^{89} in larger fallout particle size fractions which generally represents most of the fallout radioactivity in areas close to Ground Zero. This fractionation of Sr^{89} and Sr^{90} with respect to particle size may be predicted on the basis of the different half-lives of their noble gas precursors, Kr^{89} and Kr^{90}, respectively, and the rate of particle formation.

A summary of the number of detonations at NTS from 1951 to 1959, their yield, and type of support during detonation is given in Table 2.

TABLE 2

Surface and above surface nuclear detonations at NTS

	Tower Mounted		Air Drops and Balloon Mounted		Surface and Sub-Surface		Total
Year	No.	KT Yield*	No.	KT Yield*	No.	KT Yield*	Detonations
1951	1	<0.1	9	110.2	2	2.4	12
1952	4	51.8	4	52.2	0	---	8
1953	7	138.7	4	113.1	0	---	11
1955	10	158.3	3	7.7	1	1.1	14
1957	9	140.4	13	199.6	1	0.3	23
1958	5	0.2	11	18.3	4	19.2	20
Total	36	489.5	44	501.1	8	23.0	88

*Reference (4)

The deposition of fallout from these devices has ranged from almost background levels after aerial bursts to 25% of the radioactivity created by a tower-supported weapon. Based on approximately 1g of Sr^{89} or Sr^{90}/KT, our estimates of the Sr^{89} and Sr^{90} deposited from these NTS detonations between 1 mi. from Ground Zero and H + 12 hrs. are shown in Tables 3 and 4 respectively.

TABLE 3

Estimated distribution of Sr^{89} from surface and above surface detonations at NTS

	Estimated Total Curies of Sr^{89} Produced	Curies of Sr^{89} Deposited by H + 12 hrs.	Curies of Sr^{89} Available for Deposition Elsewhere
Tower Mounted	13,570,000	271,500	13,298,000
Balloon and Air Drops	13,780,000	13,800	13,766,200
Total	27,350,000	285,300(1%)	27,064,200

TABLE 4

Estimated distribution of Sr^{90} from surface and above surface detonations at NTS

	Estimated Total Curies of Sr^{90} Produced	Curies of Sr^{90} Deposited by H + 12 hrs.	Curies of Sr^{90} Available for Deposition Elsewhere
Tower Mounted	80,278	6,422	73,856
Balloon and Air Drops	87,180	1,644	80,536
Total	162,458	8,066	154,397

Therefore, one can speculate that if the Sr^{90} which is not deposited by H + 12 hrs. were uniformly deposited over the United States the average could be approximately 50 mc/mi.^{2}. In comparison the average Sr^{90} calculated from 1959 soil data(5) was 62 mc Sr^{90}/mi.^{2} on the U. S. mainland. The similarity between these Sr^{90} values suggests fallout from NTS may be a substantial contributor to soil Sr^{90} found at distances beyond 12 hrs. cloud travel from NTS.

Particle Size Distribution in Fallout Patterns

The size of deposited fallout particles decreased with greater distance from Ground Zero. Also, the size of fallout particles decreased with greater lateral distance from the line of maximum radiation intensity along a fallout pattern or "midline" of fallout[1,3,6]. The relative amount of β radioactivity associated with the particle size fraction less than 44μ in diameter was increased by reduction in the mass of the device support and of cab materials; therefore, the amount of radioactive fallout deposited at greater distances from Ground Zero increased in this particle size fraction.

It was found that leaf material from vegetation in the environs of the Nevada Test Site during Operation TEAPOT (1955) retained predominantly the less than 44 μ diameter fallout particles.[7] Therefore, this size range has been emphasized in our more recent studies.

Within the limits of 1 mi. from Ground Zero out to a distance corresponding to H + 12 hrs. fallout time, radioactive fallout deposited from test devices detonated on 500- and 700-ft. towers had approximately 30% of the radioactivity in the less than 44μ diameter particle size range. However, a test device of nearly comparable yield mounted on a 700-ft. balloon had 70% of its deposited radioactivity in the less than 44μ particle size range.

Within the less than 44μ diameter particle size range samples, there was from 38 to 50% of the radioactivity associated with particles less than 5μ in diameter in fallout from tower-supported detonations; from 51 to 83% of the radioactivity was associated with the less than 5μ diameter particles from balloon-supported detonations. Significant percentage contributions of radioactivity by particles less than 5μ in diameter were observed at virtually all sampling locations for both tower and balloon-supported detonations.[1]

Other Radiochemical Properties of Fallout Debris

Fallout particles less than 44 μ in diameter had greater percentages of Sr^{89}, Sr^{90}, and Ru^{103}, Ru^{106} at 30 days after detonation than did the larger sized particles. The percentages of Sr^{89}, Sr^{90} and Ru^{103} and Ru^{106} in fallout debris from balloon-supported detonations were 2 to 4 times higher than in corresponding particle sizes from tower-supported detonations. The reverse was observed for Zr^{95}. Ba^{140}, Ce^{141}, Ce^{144} and Y^{91} varied to a lesser degree between fallout from tower and balloon-supported detonations. Sr^{90} averaged 2.7% of the total radiostrontium at D + 30 days in fallout from detonations supported by towers.[1]

Solubility of Fallout Debris

Solubility of fission products from fallout debris is one of the most important properties to consider with respect to the "internal emitter"

of biological systems. The solubility of fission products from fallout debris in water and in 0.1 N HCl has been used by this laboratory arbitrarily as indices to biological availability.

The radioactivity in fallout debris from tower-supported detonations has been observed to be from 1 to 2% soluble in water.[8,1] Fallout debris from balloon-supported detonations was more soluble in both water and 0.1 N HCl than that produced by other types of detonations.[1] The solubility of fallout debris from tower-supported detonations increased with decreasing particle size; however, in the case of balloon-supported detonations, the smaller sized particles were somewhat less soluble than larger particles (Table 5).

TABLE 5

Solubility in water and 0.1 N HCl of fallout debris from tower and balloon-supported detonations

Support	Particle Size Range (μ)	Solubility (Per cent of β Activity)	
		Water	0.1 N HCl
Tower	>44	<1	5
	<44	<2	14 to 36
Balloon	>44	31	>90
	<44	14	>60

Radioactivity Ratios of Cs^{137} and Sr^{90}

The percentages of Sr^{90} recovered from soil after leaching with 6 N HCl varied from 13 to 100% among samples taken to 6 in. depth from 7 fields of alfalfa in the NTS area. Ratios of Cs^{137} to Sr^{90} ranged from 1.57 to 11.7 when calculated from these Sr^{90} determinations and gamma spectrometry Cs^{137}. The range of Sr^{90} solubilities, and the corresponding range in radioactivity ratios, precluded acceptance of acid extraction for assessing the levels of Sr^{90} in NTS soils. Therefore, in the following discussion of Sr^{90} in soils all the values were determined by the Na_2CO_3 fusion method of analysis.

An interest in rapid survey procedures for Sr^{90} led to limited investigation of Cs^{137}/Sr^{90} ratios for predicting Sr^{90} levels. The ratios found in soils near NTS averaged 1.55 ± 0.79 in 1957 and 1.33 ± 0.13 in the McCook, Nebraska, area in 1960. (Table 6).

TABLE 6

Cs^{137}/Sr^{90} ratios in soils samples
from the NTS and McCook, Nebraska, Areas

NTS Location (Alfalfa Fields)	Cs^{137}/Sr^{90}* Oct. 1957	McCook location (Fence Lines)	Cs^{137}/Sr^{90}* Sept. 1960
Alamo, Nev.	2.68	Haigler, Nev.	2.18
Beaver, Utah	2.45	Sharon Spgs, Kan.	1.24
Caliente, Nev.	1.69	Atwood, Kan.	1.24
Mesquite, Nev.	1.55	Oakley, Kan. (10 mi. N)	1.16
Cedar City, Utah	0.92	Oakley, Kan. (2.9 mi. N)	1.13
Lund, Nev.	0.91	Smoky Hill	1.07
St. George, Utah	0.66		
NTS mean (1957)	1.55 ± 0.79**	McCook mean (1960	1.34 ± 0.13**

* Sr^{90} determined after fusion of the soil in Na_2CO_3.
** Standard deviation

The variation among the individual Cs^{137}/Sr^{90} ratios of the NTS soils shows the predictions of soil Sr^{90}, from soil Cs^{137}, can differ by a factor of 2 from what is present. The precision in predicting Sr^{90} appears to increase with distance from detonation sites. This is shown by the much smaller standard deviation of the ratios for the midwestern soils and by that reported by Gustafson for a much larger geographical area beyond 300 mi. from NTS.(9)

Our empirical Cs^{137}/Sr^{90} activity ratios in soil from the NTS area differ appreciably from point to point and from the theoretical 1.8(9,10). In general, the ratio decreased as the deposited fallout increased(Table 7).

TABLE 7

Sr^{90} in soil and associated Cs^{137}/Sr^{90} ratios near NTS in October, 1957

Location (Soil 0-6 in. deep)	Soil Sr^{90} $mc/mi.^2$	Cs^{137}/Sr^{90} Activity Ratio
Alamo, Nev	30.8	2.68*
Beaver, Utah	28.8	2.45
Caliente, Nev.	22.9	1.69
Mesquite, Nev.	39.1	1.55
Cedar City, Utah	62.4	0.92
Lund, Nev.	64.4	0.91
St. George, Utah	111	0.66
Tempiute, Utah	218**	0.31

* Cs^{137} determinations made at Argonne National Laboratory by Dr. P. F. Gustafson

** Virgin soil 0-2 in. deep

The heaviest depositions of local fallout in NTS areas came from devices detonated on towers. The radioactivity ratios of particles from one such device are shown in Table 8.

TABLE 8

Cs^{137}/Sr^{90} ratios of Smoky fallout particles

Particle Diameters (μ)	Hour of Fallout Arrival			
	H + 1.15	H + 4.5	H + 5.6	H + 6.5
0 - 44	0.77	0.47	0.36	0.35
44 - 88	0.37	0.36	0.42	0.30
149 - 250	0.20	---	---	0.27

These ratios are 10 to 40% of the theoretical ratios and show that the particles are Cs^{137}-deficient relative to Sr^{90}. Analyses of local soil samples and NTS produced fallout particles indicate dependence upon theoretical nuclide ratios for predicting Sr^{90} from Cs^{137} contents may be misleading in the case of "close-in" fallout from individual detonations.

Milk samples were collected at various times in 1957 for Sr^{90} and Cs^{137} determinations. The ranches or farms sampled were located between Milford, Utah and Bishop, Calif. The average Cs^{137}/Sr^{90} ratio for milk produced in late April (pre-PLUMBBOB) was 5.40 ± 1.64 compared to 6.15 ± 2.77 in October, after the test series.

Milk was collected 8 to 57 days after deposition of fallout onto farms in fallout patterns with > 2 mr/hr. at H + 12 hrs. during Operation PLUMBBOB. The best fitting relationship between Sr^{90} and Cs^{137} in 16 samples was an exponential curve derived by least squares:

$$\mu\mu c\ Cs^{137}/l = 12.4\ (\mu\mu c\ Sr^{90}/l)^{0.46}$$

The activity ratios found in milk were lower than those reported for the continental U. S. in 1958 samples.(9) The mean ratio for 15 samples collected near NTS in August 1959 was 5.14 ± 2.29. The coefficients of variation for these groups of milk samples ranged from 30% to 45%. This is higher than the 22% reported by Gustafson. The large variation found in activity ratios of milk samples was sustained nearly a year after direct contamination of feeds by "local" fallout ceased.

Apparent Environmental Equilibrium

In 1958 and 1959, jack rabbits were collected from Yucca Flat itself to a distance of over 400 mi. While most of these areas were contaminated during the Operation PLUMBBOB, most (if not all) were contaminated to varying degrees during previous test series.

The Sr^{90} soil levels in 1958 and the Sr^{90} rabbit bone levels in 1958 and 1959 for the various sampling sites are listed in Table 9.

TABLE 9

1958 Sr^{90} soil levels and 1958-1959 jack rabbit bone levels at various sampling sites in Nevada and Utah

Approx. mi. from NTS	Sampling Area	1958 Soil Sr^{90} mc/mi.^{2}	Bone ($\mu\mu$c Sr^{90}/g Ca)* 1958	1959
0	Smoky Tower	9014	50.4	43.2
20	Area I, Nev.	513	19.0	64.4
74	Moapa, Nev.	16	15.8	18.0
76	Delamar, Nev.	23	14.6	17.1
80	Warm Springs, Nev.	93	26.8	30.0
82	Glen Rox, Nev.	142	21.8	19.6
96	Overton, Nev.	21	15.5	13.9
132	Belmont, Nev.	32	23.8	28.0
135	St. George, Utah	46	19.6	25.3
136	Enterprise, Utah	41	13.7	19.3
232	Clear Lake,Utah	26	11.1	18.5
235	Antimony, Utah	29	16.2	17.3
240	Antimony-Otter Cr., Utah	44	15.0	15.4
270	Fremont, Utah	26	17.4	14.2
272	Reno-Sparks, Nev.	16	27.3	19.0
300	Fountain Green, Utah	38	13.3	22.4
356	Columbia, Utah	67	20.6	20.7
432	Vernal, Utah	14	11.9	12.9

*"Strontium Units"

The data suggest two observations: first, the highest bone levels are frequently associated with the higher soil Sr^{90} levels but the relationship between bone and soil Sr^{90} levels is not linear. Second, the bone levels remained essentially unchanged over the 1958-1959 period with the increases matched by the decreases.

The persistence of Sr^{90} in the soil environment was examined by detailed studies conducted in the PVI area, 99 mi. from Ground Zero, and in a second area, PVII, 136 mi. from Ground Zero, both in the Smoky fallout pattern. The results are summarized in Table 10, and indicate that within the limits of sampling accuracy, the Sr^{90} surface soil levels were unchanged over the 12-month period.

TABLE 10

Effect of time on Sr^{90} surface soil (0-1") levels

Area	Distance from NTS	Soil Sr^{90} level of Samples Collected at D + 3 days	D + 12 months
		$mc/mi.^2$ at time of analysis (1959)	
PVI	99 mi.	127 ± 15	109 ± 31
PVII	136 mi.	95 ± 15	114 ± 19

Samples of the soil profile were collected in the PVI area 3 days, 12 months, and 24 months after the Smoky shot. The analysis of these samples (Table 11) indicates that the Sr^{90} is primarily restricted to the surface inch with relatively small amounts in the second inch, which is in agreement with similar studies of gross β radioactivity and Pu movements over considerably longer periods of time in New Mexico soils contaminated by the Operation TRINITY shot of 1945.(11)

TABLE 11

Effect of time on distribution of Sr^{90} in soil profile, Area PVI

		Soil Sr^{90} Level of Samples Collected at		
		D + 3 days	D + 12 mo.	D + 24 mo.
	Depth (in.)	mc/mi.^{2} at time of analysis (1959)		
Stake 1	0 - 1	104	89.5	128
	1 - 2	19.2	13.7	9.20
	2 - 3	Bkg	Bkg	--
	3 - 4	Bkg	Bkg	--
Stake 13	0 - 1	112	154	106
	1 - 2	22.9	9.76	14.2
	2 - 3	15.9	Bkg	--
	3 - 4	2.79	Bkg	--
Stake 16	0 - 1	130	169	178
	1 - 2	19.5	29.8	3.07
	2 - 3	Bkg	5.58	--
	3 - 4	Bkg	Bkg	--

These studies, as well as the relatively unchanging Sr^{90} levels over the 1956 to 1960 period in Area I, supports the concept of a persistent Sr^{90} soil contamination.

Biological Availability of Fallout Sr^{90}

Studies involving kangaroo rats and jack rabbits as native biological indicators were conducted in NTS fallout areas during Operation UPSHOT/KNOTHOLE in 1953(12), Operation TEAPOT in 1955(7), and Operation PLUMBBOB in 1957.(1)

Differences in the half-lives of the precursors suggest that the distribution of Sr^{89} should not be necessarily indicative of the distribution of Sr^{90}. The proportion of Sr^{89}/Sr^{90} in the bone ash of jack rabbits along the midline of a TEAPOT fallout pattern was found highly variable at different distances from Ground Zero.(13)

Subsequent studies were specifically related to the distribution of Sr^{90} in the environment and its accumulation by small native animals. Because of the chronic nature of Sr^{90} contamination, the time sequence of

accumulation has been emphasized.

A study area, Area I, was established in 1952 approximately 13 mi. north of NTS following the fallout from Operation JANGLE in November 1951. As indicated in Table 12, soil Sr^{90} levels in this area were increased by approximately an order of magnitude with this event.

TABLE 12

Soil and jack rabbit bone Sr^{90} levels in Area I, 13 mi. from NTS

Contamination Event	Date of Collection	Soil Sr^{90} (mc/mi.^{2})	Bone Sr^{90} ($\mu\mu$ c/g Ca)
RANGER Jan./Feb.1951	Sept. 1951	23*	---
BUSTER/JANGLE Oct./Nov. 1951	Nov. 1951	200**	---
	Oct. 1952	---	33 ± 13
UPSHOT/KNOTHOLE Mar./June 1953	July 1953	438 ± 56	---
	Apr. 1954	---	26.3 ± 12.5
TEAPOT Feb./May 1955	Apr. 1955	---	9.4
	Oct. 1955		11.8 ± 6.3
	Oct. 1956	570 105	11.0 ± 0.85
PLUMBBOB May/Oct. 1957	June 1957	---	24.1 ± 6.9
	Aug. 1957	---	25.7 ± 12.4
	Aug. 1958	560 ± 73	19.0 ± 1.3
HARDTACK II Sept./Oct. 1958			
KIWI A July 1, 1959	Aug. 1959	386 ± 87	64.4 ± 33.8
	May 1960	564 ± 95	22.8 ± 12.7

* 0.9 mi. northwest of area
** Estimated on basis of contaminated soil flats located pre-shot.(14)

The biological availability of this fallout to plants in glasshouse studies was considerably greater than that observed for tower-shot fallout of which subsequent fallouts consisted--at least until Operation PLUMBBOB in 1957.(15) This material plus an increment of fallout from Operation TUMBLER/SNAPPER resulted in a value of 33$\mu\mu$ c Sr^{90}/g Ca in the bone of rabbits collected in the fall of 1952, the highest value observed until 1959.

No apparent increase in Sr^{90} of bone was recorded following Operation UPSHOT/KNOTHOLE in the spring of 1953 nor following an apparent 30% increase in soil Sr^{90} during TEAPOT in 1955. An approximate doubling of the previous year's bone level was detected during the PLUMBBOB in 1957, although soil levels apparently were not increased. An increase was also observed immediately following the first nuclear propulsion experiment (Project ROVER, Kiwi A) in 1959.

During Operation PLUMBBOB in 1957, Area PVI, was established 99 mi. from Ground Zero on the midline of the Smoky shot fallout pattern. The Smoky shot increased the Sr^{90} contamination level of Area PVI by an estimated 65% to approximately 100 mc/mi.^{2}. Kangaroo rats and jack rabbits were collected 3, 5, 9, 13 and 20 days after fallout to determine the early rates of fission product accumulation; as shown in Table 13, kangaroo rats and jack rabbits showed an early response to the additional Sr^{90} increment, at least as early as the fifth day. A rapid equilibrium was also demonstrated between the animals and the environment since maxima were generally reached well before the 20-day sampling period was completed. Similar rapid response and equilibration with the environment were observed with respect to the shorter-lived bone-seeking fission products Sr^{89} and Ba^{140} in rabbit bone after Smoky and other detonations during PLUMBBOB.(1)

TABLE 13

Sr^{90} levels of kangaroo rat and jack rabbit bone in Area PVI, 99 mi. from Ground Zero

Date of Collection	μμ c Sr^{90}/g Ca in Bone	
	Kangaroo Rat	Jack Rabbit
Oct. 12/55	---	20.6
Aug. 31, 1957 Contaminating Event: Smoky Shot		
Sept. 3/57	5.34	20.7± 9.93
Sept. 5/57	6.62	22.7
Sept. 9/57	6.43	26.8± 13.6
Sept. 13/57	9.64	25.0± 6.98
Sept. 20/57	8.69	----
July 3/58	8.33	----
July 7/58	8.48	19.4± 5.27
Aug. 12/59	---	33.4± 12.7
May 1960	---	19.3± 7.04
May 1961	---	10.0± 6.32

Subsequent sampling of kangaroo rats showed that the maximum levels of Sr^{90} bone contamination reached during the 20-day initial sampling period tended to be maintained one year later. Jack rabbit bone levels reflected an increase in 1959, a return to the 1958 level in 1960, and an abrupt drop in 1961.

The relatively poor correlation between soil and bone Sr^{90} levels, suggests that some fraction of the Sr^{90} fallout, rather than the total, is of primary significance with respect to biological uptake. For example, when 1958 soil samples collected along the Smoky midline were leached with 6 N HCl, the correlation between the Sr^{90} soil levels so obtained and the corresponding bone levels was much improved (Table 14). It is

TABLE 14

Comparison of bone Sr^{90} levels to soil Sr^{90} levels determined by fusion and 6 N HCl extraction (1958 sample collection)

Location	Mi. from NTS	Jack Rabbit bone ($\mu\mu$c Sr^{90}/gCa)	Total Soil Sr^{90}*	HCl Soluble Sr^{90}**	Percent Soluble Sr^{90}
			mc/mi.^{2}		
Smoky Area	<1	50.4	9014	980	10.9
Nye 1, Nev.	12	23.2	933	58	6.2
Glen Rox, Nev.	80	21.5	142	18	13.0
Enterprise, Utah	140	13.8	41	27	66.7
Panguitch, Utah	205	12.9	32	16	50.3
Columbia, Utah	350	20.9	67	48	71.8

* Determined by Na_2CO_3 fusion method of analysis.
** Determined by extracting with 6 N HCl.

noteworthy that the Sr^{90} which is soluble in HCl represents an increasing percentage of the total Sr^{90} as distance from Ground Zero is increased. which, of course, agrees well with the increasing percentage contribution of small fallout particles at greater distances from Ground Zero. Equally important is the fact that during PLUMBBOB fallout materials less than

44μ in diameter were identified in respectable quantities at virtually all sampling locations within every fallout pattern, and as close as 5 mi. from Ground Zero.(1) In addition to enhanced solubility properties over larger materials, such particles are somewhat enriched in Sr^{90} content due to a rare gas precursor which limits the incorporation of this particular nuclide chain at the earlier time of formation of larger particles. Consequently, it seems feasible to consider the more soluble and ubiquitous small fallout particles as the major source of Sr^{90} to the native animals in the fallout pattern.

Despite what appears to be a rather constant Sr^{90} soil environment, sharp drops in the jack rabbit bone levels of Sr^{90} were observed in Area I in 1955 and in Area PVI in 1961. If the bone levels of jack rabbits collected in the latter area in 1958 and 1961 are plotted as a function of body weight, which may be used as a rough indication of animal age, the values are distributed as illustrated in Fig. 1. In 1958, one year after the Smoky shot contamination, 41 of 43 animals had Sr^{90} bone levels in excess of 10 μμ c/g Ca regardless of weight (or age). In contrast, in 1961, 39 of 53 animals had bone levels less than 10μμ c Sr^{90}/g Ca. The higher levels were restricted to the heavier, or older animals.

The fact that older animals of the 1961 population included individuals having both low and high Sr^{90} levels, coupled with an estimated average life span of 3.2 yrs. for jack rabbits in this area(16) strongly implies that the higher levels of Sr^{90} are associated with animals which were living early in the sequence of contamination--that is, the 1957 PLUMBBOB fallout--rather than with animals which were born later and merely lived in a contaminated environment.

There are a number of possible explanations for a lower Sr^{90} bone level of animals born subsequent to contaminating events. For example, plant foliage serving as food has a higher contamination level immediately after fallout. It was shown during TEAPOT in 1955 that vegetation tends to be a selective fallout collector for fallout particles which range predominantly from < 1 to 14 μ in diameter in the great number of samples investigated.(1) During PLUMBBOB it was additionally shown by comparing the decay rate of fallout in the environment to the β radioactivity level of serially-sampled plant leaves, that the entrapped particles were persistent for the sampling period extending from 3 to 20 days after fallout. However, the contamination of new growth appears to be due to a very few particles of redistributed fallout. Gross β analysis and autoradiograms of PVI plant material collected in 1961 did not reveal the presence of particulate contamination. It should also be noted that native vegetation in Nevada has not been shown to accumulate fallout-derived fission products via the root system; autoradiograms of plant materials collected during or immediately after test series have invariably revealed point-source contamination indicative of particulates rather than the diffuse distribution pattern indicative of metabolized radionuclides.

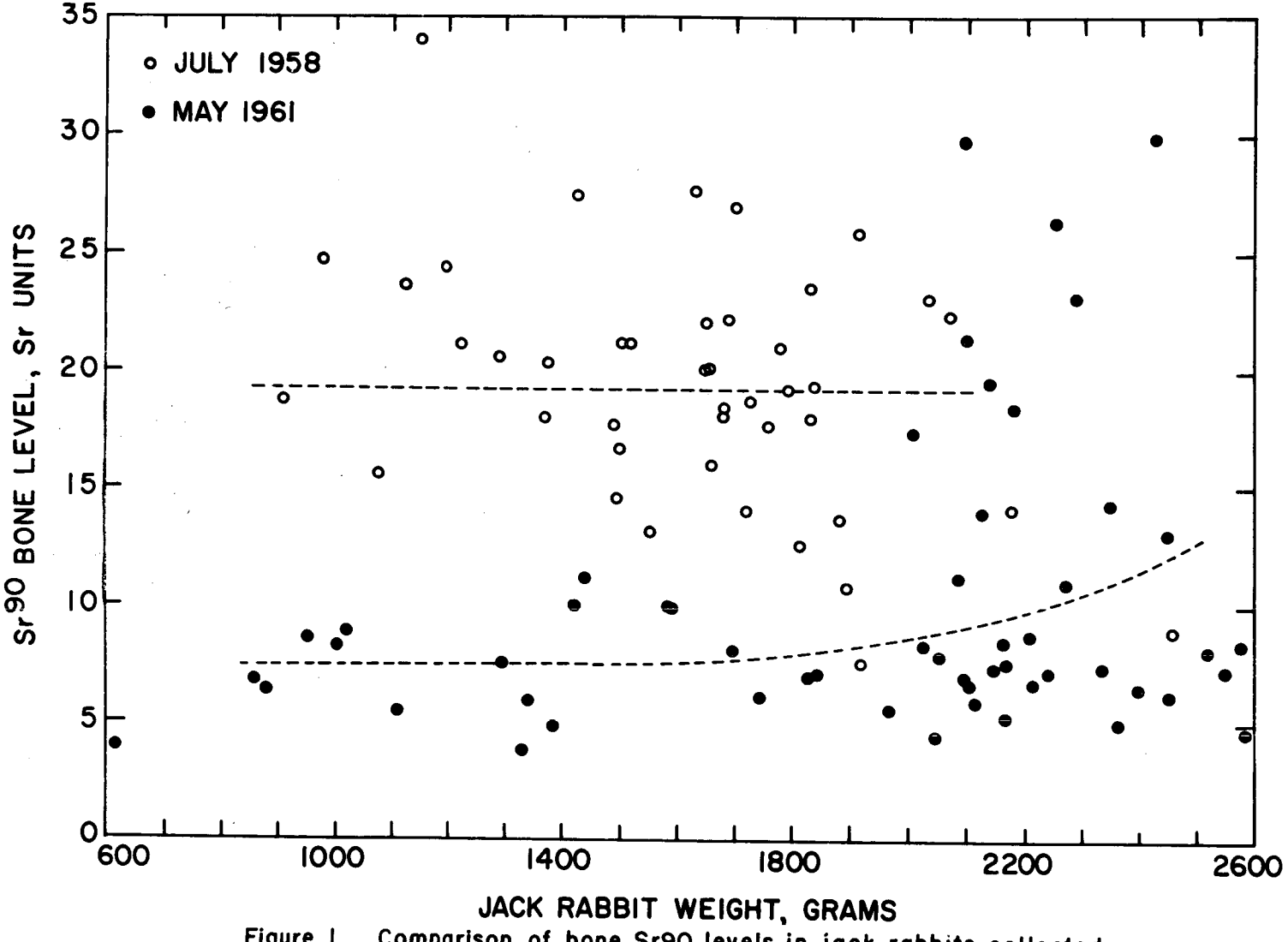

Figure 1. Comparison of bone Sr90 levels in jack rabbits collected near PVI Area in 1958 and 1961

Several observations among the data presented lend support to the concept of an available component to native animals of the total Sr^{90} contaminating the respective areas shortly after fallout. In Area I (Table 12) the June 1957 increase of Sr^{90} in bone was approximately twice the 1956 level and had occurred within a few days after the first detonation of PLUMBBOB. The July 1959 increase of Sr^{90} levels in bone in the same area immediately followed the Kiwi A experiment adjacent Jackass Flats, where similar abrupt increases occurred. In Area PVI (Table 13) the maxima in bone levels reached rapidly during the 20-day serial sampling period likewise suggest a readily available component. Under such circumstances, inhalation as a mechanism might be suspect; however, inhalation was shown to be relatively unimportant as a method of particulate contamination by restrained domestic rabbits during TEAPOT in 1955.(17)

The relative importance of the various pathways by which fallout derived Sr^{90} may enter the animal is not readily apparent. It is apparent, however, from the occurrence of reductions in the Sr^{90} bone levels of the jack rabbit population several years after contaminating events that the biological availability of Sr^{90} is much greater at some early time after fallout; it is also quite likely that mechanisms for the reduction of potentially available Sr^{90} exist in the environment, regardless of apparently persistent Sr^{90} soil levels.

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FRACTIONATION IN SURFACE BURSTS

Edward C. Freiling
U. S. Naval Radiological Defense Laboratory
San Francisco, California

Abstract

A description is presented of a combination of A. D. Anderson's Dynamic Fallout Model (NRDL-D Model) with C. F. Miller's thermodynamic model of fractionation which will be used to account for fractionation in fallout predictions. Weaknesses of the method are pointed out. Data required for testing the model (both from laboratory experiments and nuclear detonations), and for achieving more meaningful documentation of future nuclear bursts, are discussed.

Introduction

At NRDL* we are carrying out a project supported by the Atomic Energy Commission under the general supervision of the Fallout Studies Branch of the Division of Biology and Medicine entitled: "The Formation, Distribution and Characteristics of Nuclear Debris." The objective of this project is to develop a generalized fallout model capable of quantitatively predicting the formation and composition of nuclear bomb debris and its partition between stratospheric, tropospheric and local fallout. We are carrying out this project in two concurrent phases. The first phase is designed to produce short-range results and the second phase is oriented towards long-term progress. The present paper is concerned with the short-range phase and the following paper (page 47) on air

* U. S. Naval Radiological Defense Laboratory, San Francisco, California.

burst fractionation is concerned with the long-range phase. We are fortunate in having the opportunity to describe this work at such an early stage of development and the concomitant advantage of conferees' comments, which we earnestly solicit.

In the present paper I will describe our efforts to establish a computer program to calculate fallout patterns which account for the phenomena of fractionation. Fractionation is defined as any process which causes the radionuclide composition of a sample of nuclear debris to be unrepresentative of the debris taken as a whole, and the subject has been treated in detail elsewhere.(1) The manner of accounting for fractionation consists essentially of a union of the NRDL dynamic fallout model of Anderson(2-5) with the thermodynamic model of fractionation devised by Miller.*

This paper is divided into five parts. Part 1 describes the fractionation model, Part 2 describes the fallout model, Part 3 describes the adaptation of the fractionation model to the fallout model, Part 4 discusses means of testing the results, and Part 5 describes data relating to this part of the program which should be obtained in the event that future land surface bursts occur.

It is intended to devise the computer program in a compartmented fashion in order that the various parts can be revised as new developments in theory and experimental data become available. Revision is a continuing process and the description given here may well be partially obsolete by the time of publication.

Part I. The Miller Thermodynamic Model of Fractionation

The Miller thermodynamic model of fractionation can be outlined briefly as follows: First, there is described an idealized carrier material with thermodynamic and chemical properties similar to the soil which is to be drawn into the fireball, but idealized to the point where there is no chemical interaction between the carrier material and any of the radioactive products of the detonation. Secondly, the energy of the nuclear detonation is accounted for in as reasonable a manner as possible and the fraction of that energy which goes into vaporizing and melting the idealized carrier material is calculated. When the vaporized soil condenses, it and the melted soil are considered as one single phase in equilibrium with a vapor phase. The vapor phase consists of air and uncondensed radionuclides. Its volume is taken as the maximum volume of the fireball and its temperature as that of the idealized carrier material at its melting point. The time at which this temperature is

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* Additional work on this model, described in USNRDL-TR-425, is being carried out by C. F. Miller.

reached is calculated from scaling laws, and the elemental distribution of the mass chains of the fission fragments and the other product nuclides at this time is determined from known and estimated fission yield and decay relationships. The distribution of each product element between the liquid and vapor phases is then calculated according to Raoult's Law. The idealized carrier than solidifies, freezing in the trapped radionuclides. This point marks the end of the first period of condensation. The second period of condensation treats the deposition of those product nuclides still in the vapor phase upon the frozen soil particles. Some suggestions were offered by Miller as to how such a deposition might be accounted for, but the actual calculations were never carried out. According to this model the fractionation is now seen to result from two sources. The first source is the separation of particles from the cloud before the second stage of condensation has occurred. These particles will contain normal quantities of refractory elements but will be depleted in volatile elements. With regard to those particles which remain in the cloud during the second stage of condensation, another type of fractionation occurs. Here those product elements which dissolved in the particles up to the time of solidification will have been distributed throughout the particles in proportion to the particle volume, whereas those elements which condensed after solidification will have distributed themselves among the particles in accordance with the available surface. In the remainder of this section the aforementioned processes will be described in greater detail and the defects of the model will be discussed in a later section. It should be stated that Miller was well aware of the inadequacies of this approach but for lack of time and suitable experimental data was unable to carry his calculations to a higher degree of refinement.

The properties chosen for the idealized carrier material are as close as practical to those of anorthoclase ($Na_2O.Al_2O_3.6SiO_2$). It is non-reactive towards the fission product elements and their oxides and dissolves the elements as stable oxides. Its melting point is 1400°C at which temperature its vapor pressure is negligible. The molecular weight is 524 and the entropy of fusion is 29 cal./mole degrees. The heat of fusion is 48,000 cal./mole, the dissociation energy is 3,370,000 cal./mole and the heat capacity is such that 200,000 cal./mole are required to raise the temperature of the soil from 298°K to its melting point.

The discussion of the partition of burst energy which follows is based upon the considerations outlined by Miller. It differs, however, to the extent to which new thermodynamic data and scaling relationships have been incorporated into the treatment since the initial appearance of his work. The calculations can be outlined as follows: First the manner in which the energy of the burst has been distributed at the time of the final temperature maximum is calculated. The energy is considered to be distributed between nuclear radiation, thermal radiation, blast and shock, and the internal energy of the fireball. The distribution is shown schematically in Fig. 1.

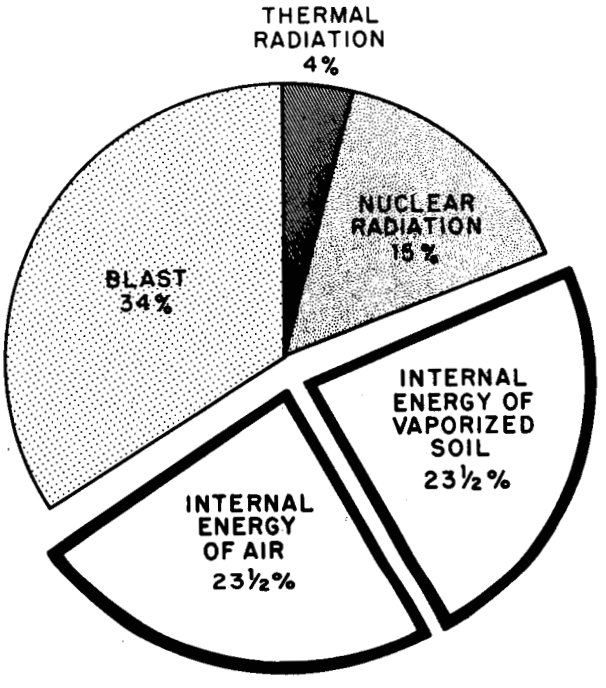


FIGURE 1. ESTIMATED ENERGY PARTITION AT THE TIME OF SECOND MAXIMUM IN MILLER'S MODEL.

In these calculations the energy equivalent of 1 KT of TNT has been taken as 10^{12} cal. The dissociation energy of air is 101,830 cal./mole. The heat capacity and pressure--volume--temperature relationship of gases have been assumed to be ideal.

Miller next defines "the model surface burst" as one in which one half of the internal energy in the fireball at the second maximum was utilized in dissociating and heating air molecules and the other half for the vaporization of soil molecules. Applying this principle leads to the result that, well within the error of estimate, equal number of moles of air and soil atoms are present in the fireball at the time of second maximum. Relating these to the total energy available by means of the perfect gas law and the estimated soil properties leads to 1.6×10^6 moles per KT of yield for each; or 840 tons of soil per KT of yield.

Hillendahl[6] gives a number of useful scaling relations for surface bursts. In his nomenclature:

W = total weapon yield (kilotons)
t = time (seconds)
ρ_0 = ambient air density (g/l)
$t^* = t/t_{fmax}$ = ratio of the time t to that of the final maximum in observed temperature

H = power (kilotons per second)
$\sigma A \psi^4$ = (Stefan's constant x total radiating area x fourth power of power temperature)
T = absolute temperature of the fireball

1 Kiloton = 10^{12} calories
R_1 = Radius of uniform brightness
R_2 = Main radiation front radius
R_3 = Absorption shell radius
R_4 = Main shock front radius
R_5 = Precursor shock front radius
(The radii are shown in Fig. 2 and are further discussed in the following paper, page 47 . They are taken in meters).

The scaling equations are

$$W = 0.65 \rho_o (2R_4)^5/t^2$$

$$t_{min} = 0.0030 W^{0.49}$$

R_4 min = R_2 min = 33 $W^{0.396}$ = the values of the radii at the minimum in observed temperature

$$H = 3.75 W^{0.51} t^{*-1.45} e^{-9e^{-2.73t^*}} e^{-93^{-12Wt^*}}$$

And from his figures one finds

$$T = 7000 W^{-0.07} t^{*-0.34}$$

while, at $t^* = 1$

$$R_1 = 48 W^{0.39}$$

$$R_2 = 66 W^{0.39}$$

$$R_3 = 75 W^{0.39}$$

$$R_4 = 90 W^{0.39}$$

From the time of second maximum to the time required to reach 1400°C. the internal energy of the cloud will decrease. Dissociated air and soil will release energy by cooling and reassociating. The energy released will be distributed among radiation, expansion, heating additional air, and heating and melting additional soil. The amount of energy radiated up to any time is obtained by integrating Hillendahl's power function. The work of expansion is calculated from the change in volume of the 1.6 x 10^6 moles per KT of air atoms due to cooling from 6000° to 1400°C. The difference between the maximum volume of the fireball calculated from Miller's scaling equation

$$R_m = 922 \times 10^3 W^{0.314}$$

and the volume of the original air at 1400°C. is used to calculate the additional amount of heated air. Some 3% of the original device energy turns out to become available for heating and melting soil. It is assumed that only half of this residual energy produces molten soil. The additional soil melted turns out to equal about half of the condensed soil. The total amount of molten soil is thus calculated to be 2150 tons for a 1.7 KT shot, in poor agreement with the 800 tons found in the Rainier shot (Nevada Test Site, 1957), where no radiation was lost, although much water was present. (cf. Part IV).

With the foregoing considerations the stage is set for the application of Raoult's Law by definition of the macroscopic state. This state consists of the calculated number of moles of molten soil in equilibrium with a vapor phase. The previous discussion took the temperature down to 1400°C., but the same principles could be used to reach definitions of the macroscopic state for intermediate temperatures. At higher temperatures, however, the vapor pressure and partial dissociation of the soil will become increasingly important.

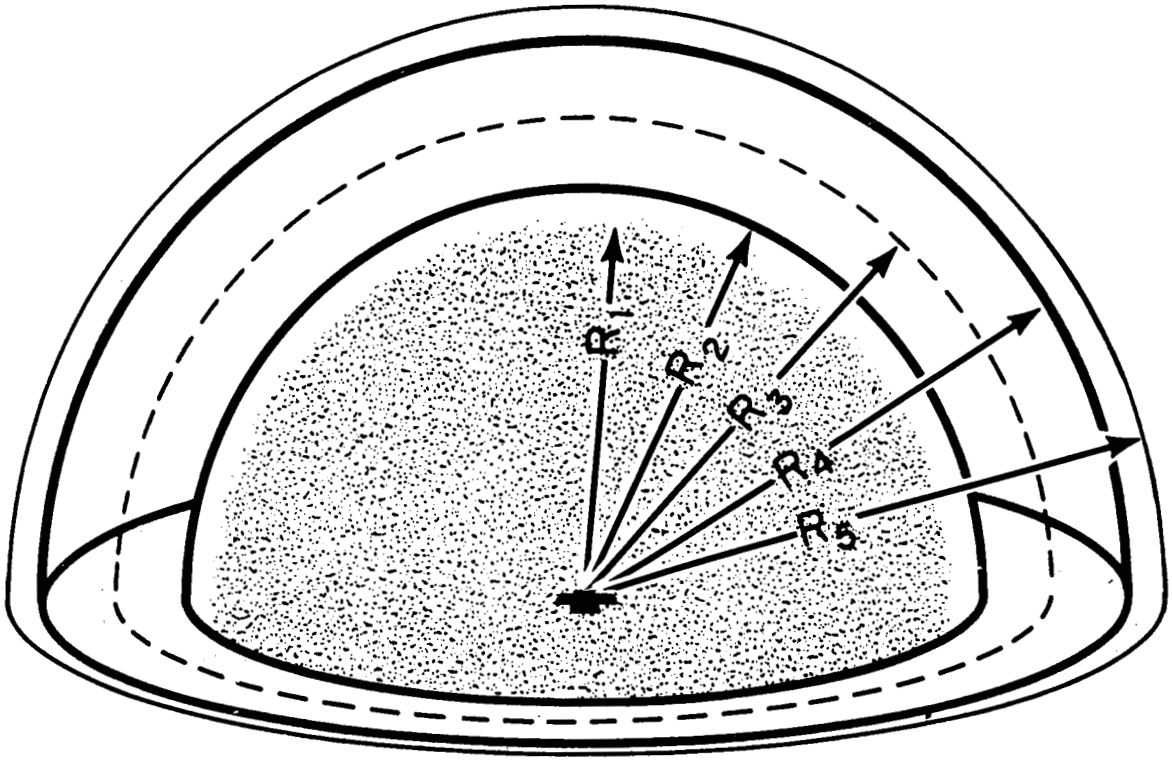


FIGURE 2. ILLUSTRATION OF FIREBALL RADII (FROM (6)).

According to Raoult's Law, the vapor pressure of each component of an ideal solution at thermodynamic equilibrium is equal to the vapor pressure of the pure substance multiplied by its mole fraction in the solution. Thus, if there are n_1 moles of component 1 and n_2 moles of component 2 in a two component solution the mole fraction of component 1 is

$$N_1 = \frac{n_1}{n_1 + n_2},$$

Its vapor pressure above the solution is

$$p_1 = N_1 p_1^{\square}$$

where $p_1^{\square}$ is the vapor pressure of the pure component. The law is illustrated graphically in Fig. 3. Here, the dashed lines illustrate ideal behavior while the solid lines indicate two cases of real behavior Positive deviations are attributed to mutual repulsion of the components, and negative deviations are attributed to mutual attraction.

In order to apply this to the system at hand, one further concept must be mentioned, namely, that of the partial pressure. In an ideal mixture of gases, the partial pressure of component is given by

$$p_1 = n_1^{\circ} RT/V$$

where n_1° is the number of moles in the vapor phase, R is the gas constant, T the absolute temperature and V the volume. Eliminating **pl** and noting that $n_1 \sim n_1/n_2$:

$$\frac{n_1^{\circ}}{n_1} = \frac{p_1^{\square} V}{n_2 RT}$$

On this basis it therefore becomes a straightforward, if complex, problem to calculate the amount of each radionuclide present, as was done by Hunter and Ballou, and then distribute each one between the vapor phase and the condensed phase. The fission product radionuclides are present in such small concentrations that interaction between them can be neglected. The values of p_i for some important elements are shown in Fig. 4

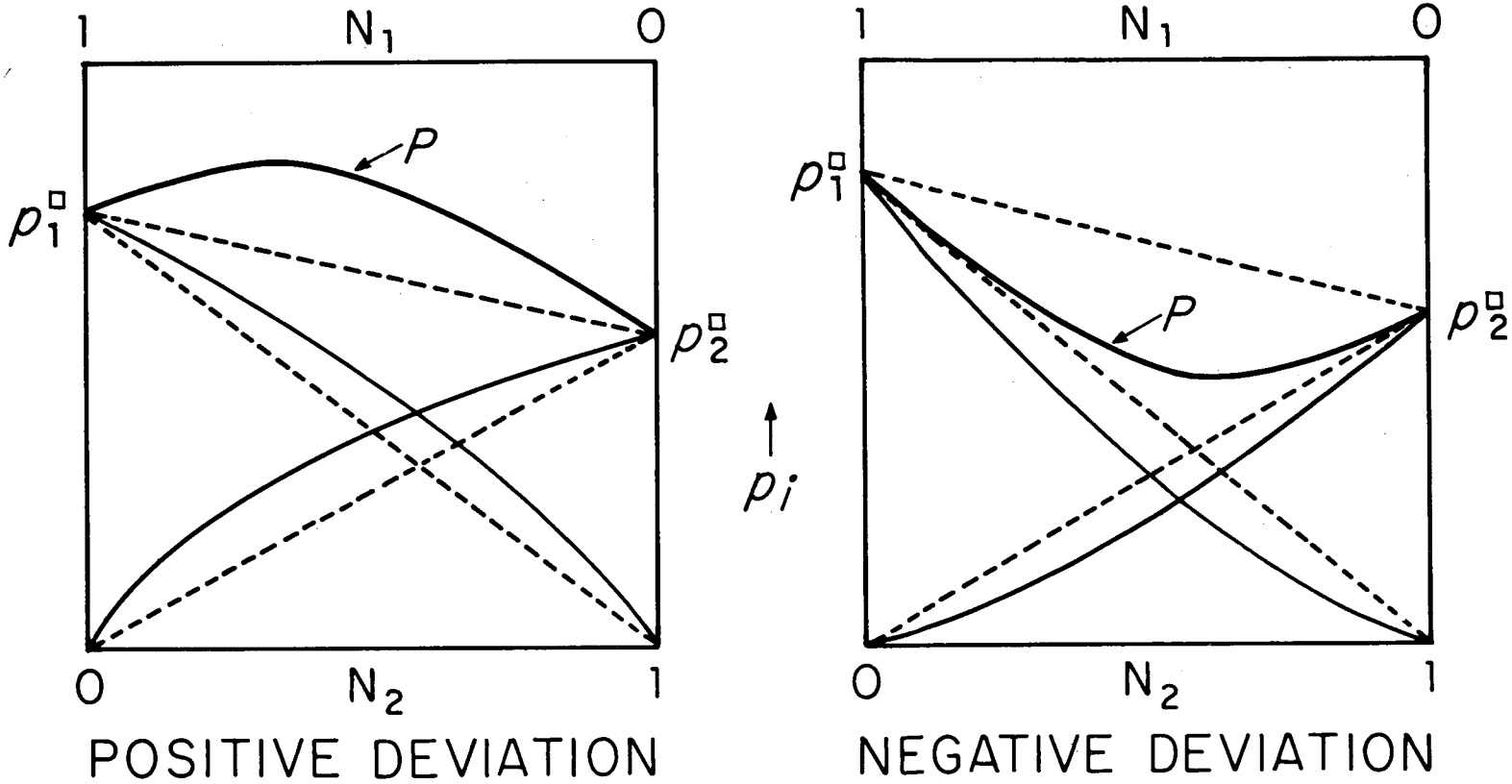


FIGURE 3. VAPOR PRESSURES OF NONIDEAL SOLUTIONS (DASHED LINES INDICATE IDEAL BEHAVIOR CALCULATED FROM RAOULT'S LAW)

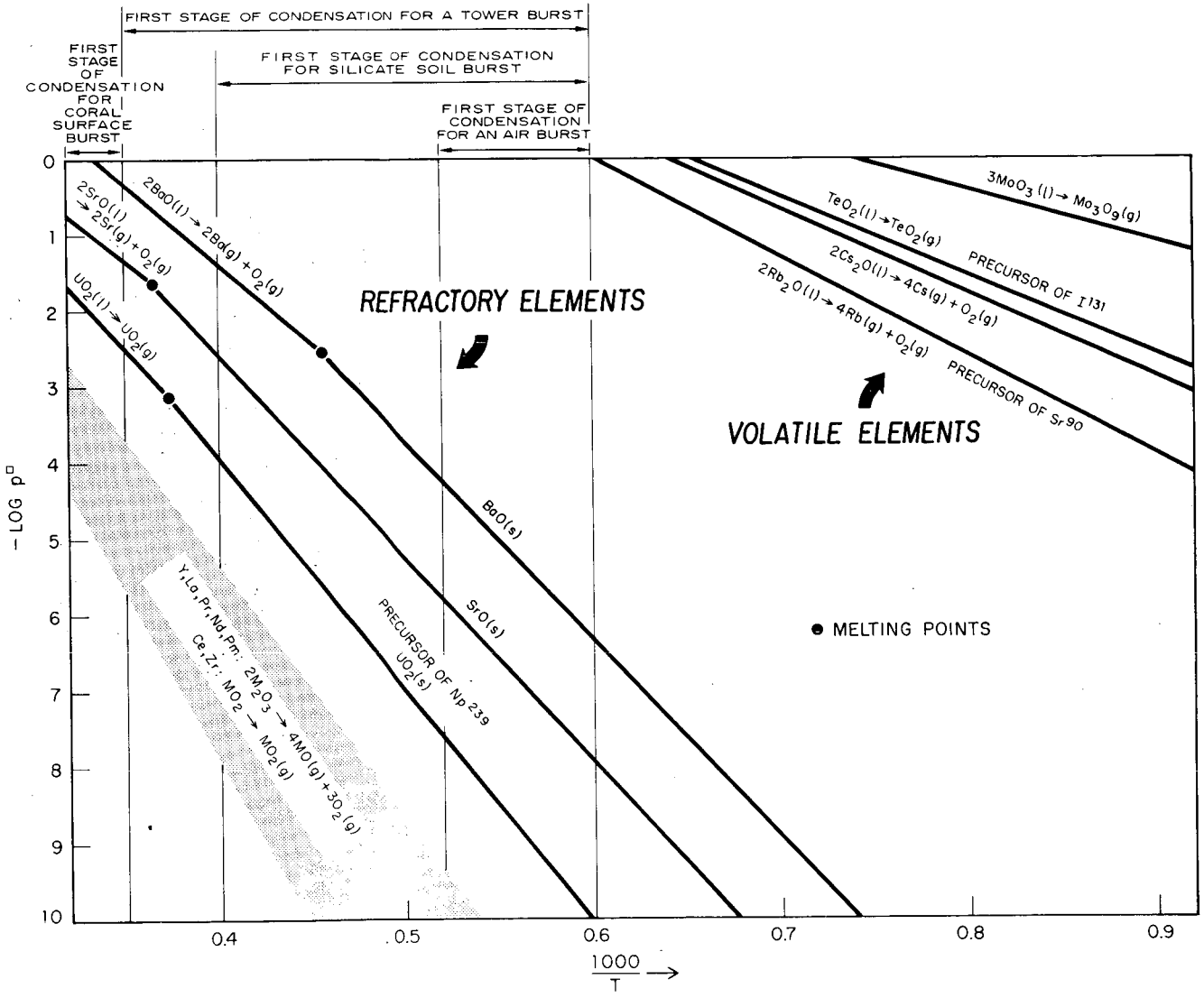


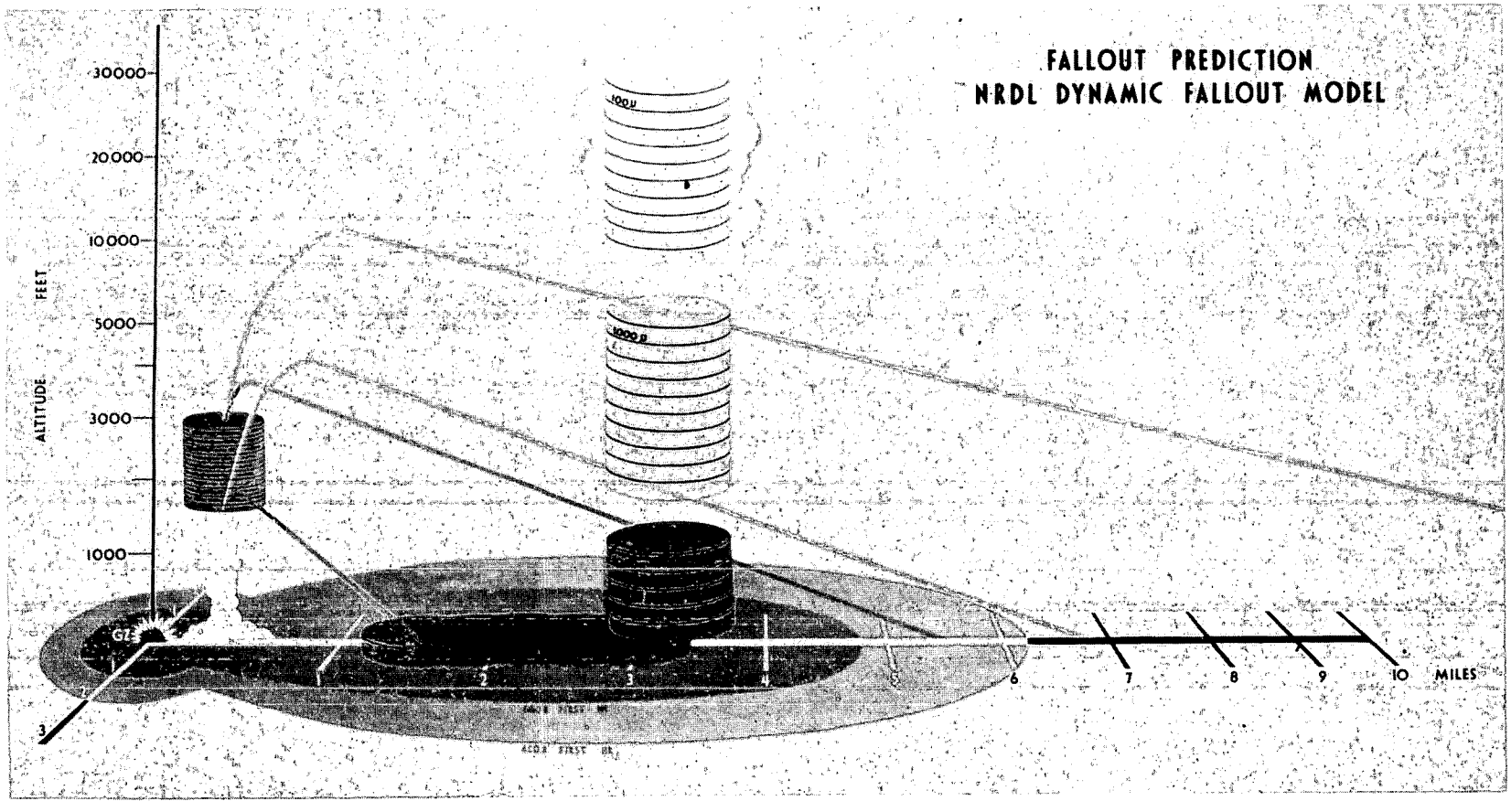
FIGURE 4. VAPOR PRESSURES OF IMPORTANT FISSION AND CAPTURE PRODUCT ELEMENTS DURING VARIOUS CONDENSATION PROCESSES.

Miller refers to the adsorption processes occurring after the particles solidify as the second stage of condensation. He suggests either using Langmuir adsorption isotherms or sublimation pressures to estimate the distributions at this stage, but doesn't apply them in his work. His results show an "ionization rate" of fractionated fission products equal to 0.4 that of unfractionated fission products at one hour, while his observed data show an "ionization rate" of 0.3 that of unfractionated fission products. From what has been said, it should be clear that there is not one fractionated composition, but many. The comparison stated is that of a single calculated composition with one of many possible observed compositions. Without radiochemical documentation the significance of the comparison remains obscure.

Part II. The NRDL Dynamic Model for Fallout from Land Surface Nuclear Bursts

The NRDL D-Model (Dynamic Model) was developed by Anderson and is described in detail in several reports.(2-5) This section will be primarily a summary of Anderson's description. Enough detail will be given to allow the reader to understand the application of Miller's fractionation model to fallout predictions for land surface bursts.

In the cooling fireball the temperature will eventually reach a point where soil particles entering the fireball are no longer vaporized and where vaporized soil present in the fireball begins to condense. This temperature is estimated as the boiling point for an abundant high boiling constituent of the soil. In the case of Nevada Test Site soil this constituent would be quartz which boils at about 2200°C. In the case of coral surface bursts this substance would be CaO which boils at about 2800°C. In the D-Model the time at which the fireball temperature reaches this value is taken as the time of the beginning of fallout. This time is about 0.6 sec. for a 1 KT burst, 6.4 sec. for a 100 KT burst, and 64.3 sec. for a 10 MT burst. The cloud at this time is approximated by a cylinder. The total residual activity is assumed to have a uniform distribution of particle sizes contained in a volume coincident with this cylinder. The activity-particle size distribution is divided into 100 classes. These classes range from a 40-60 μ diameter class up to a 8700-10,000 μ diameter class. Particles smaller than 40 μ are considered beyond the scope of local fallout. It is convenient therefore to think of the cloud at this time as consisting of 100 identical coincident right circular cylinders each corresponding to one of the particle size classes. In addition, each cylinder is divided up into N equisized discs. N may be as small as 7, as in the case of a 0.01 KT burst or as large as 231, as in the case of a 100 MT burst. The total number of discs therefore varies from 700 to 23,000. The situation is illustrated in Fig. 5.


FALLOUT PREDICTION
NRDL DYNAMIC FALLOUT MODEL
ALTITUDE FEET
30000
20000
10000
5000
3000
1000
GZ
100 Y
1000 R
1
2
3
4
6
7
8
9
10 MILES

In contrast to those fallout models which begin when the cloud has reached its maximum height, the D-Model must account for the upward motion of the cloud in calculating the vertical velocity of the particles. Thus, the rate with respect to the ground at which a particle moves upward or downward is taken as equal to the difference between the velocity with which it is carried up by the rising cloud and the velocity due to gravitational fall. Vertical winds are not considered. Wind speed and direction during the time of rise and fall determine the particles horizontal displacement. The trajectory of each disc is now taken as the trajectory of a particle whose size is the midpoint of the size class of the disc and whose location is the center of the disc. Cylinders corresponding to different particle size classes are therefore seen to diverge gradually from one another. Cylinders representing large particles will not rise very high and will fall more rapidly. Cylinders representing small size particles will rise higher and level off more slowly. The time-altitude history is approximated by a finite difference equation dividing the time intervals from the start of fallout to the time the particle reaches the ground into N smaller intervals. The first interval is 1 sec. and each interval is 1 sec. longer than the preceding interval.

In treating the stem, the D-Model ignores the much higher velocities present there and treats the motion in the same manner as for the main part of the cloud.

Fig. 6 shows the cumulative percent of residual activity for particles of different size as determined from the Operation JANGLE underground shot. It is seen to be log-normal. For most siliceous soils the sparse data available indicate that for local fallout the fraction of radioactivity in a given size range is roughly independent of yield. However, the size-radioactivity distribution for coral is markedly different from that for siliceous soil.

If one of the discs representing a particular size class lands on the ground before one hour after burst and covers a point p on a rough plane, then the deposited dose rate R in r/hr. at 1 hr. and 3 feet above this location is given by $R = 120\ FA/NS$. Here 120 is a constant that takes into account final surface irregularities and other factors. The contribution of discs arriving after one hour to the calculated one hour dose rate is accounted for. F is the fraction of total residual radioactivity associated with the particle size class of the disc. A is the total residual radioactivity in curies remaining at one hour after burst, and is taken as 5×10^8 W curies for a weapon yield which is 100% due to fission. S is the area in square feet on the ground covered by the radioactivity contained in the disc. The edge effect is ignored. The total deposit dose rate at P is the sum of the individual dose rates contributed by the discs that cover this location. The effects of induced activities and fractionation are not taken into account by Anderson. S is assumed to be πr^2 where r is the radius of the disc. The method of finding the radius of the disc is based upon the

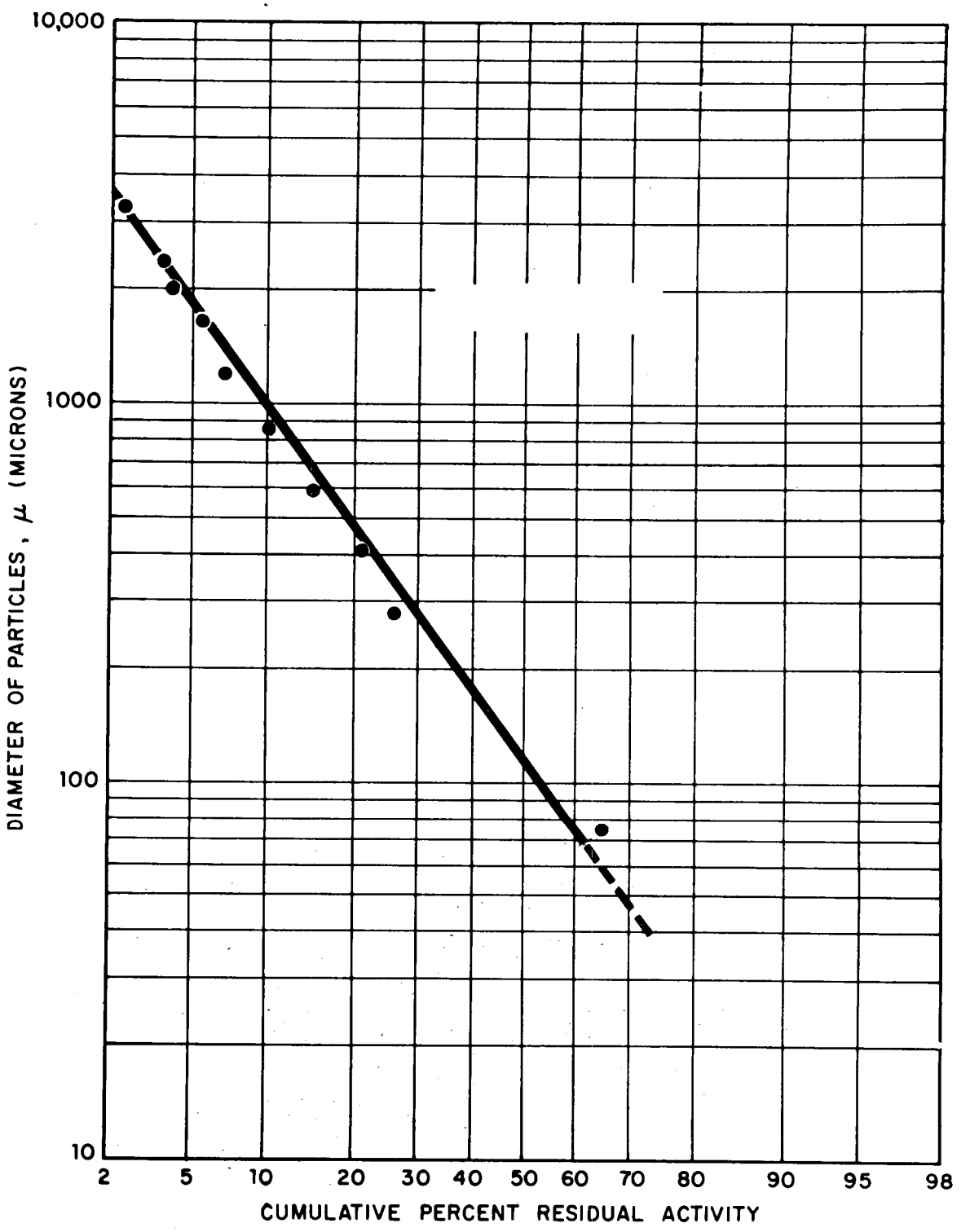


FIGURE 6. ACTIVITY-PARTICLE SIZE DISTRIBUTION FROM OPERATION JANGLE-U DATA (FROM (2)).

following three assumptions.

(1) The fallout is dispersed uniformly throughout the initial visible cloud at the time of start of fallout,

(2) As the cloud expands, the fallout expands laterally at the same rate as the cloud, and

(3) The lateral expansion of fallout stops after the cloud has stopped expanding.

The maximum disc diameter for all yields is taken as that given for 360 sec.

Because of the early time at which the D-Model begins to consider the fallout processes it can be readily seen that it is particularly suitable to being modified for the effects of fractionation according to the methods of Part I above. The details of this modification are described in the following section.

Part III. Modification of the NRDL Dynamic Model for Fractionation

The modification of the NRDL dynamic model for fractionation has been carried out by Lee, Kiley, and Johnson(7) according to a method outlined by Miller and is in the process of being programmed for an IBM 704 computer. The modification proceeds as follows. First, it is necessary to change from the activity-particle size input of Anderson to a mass-particle size input. To do this we use Miller's cratering equation

$$M = 5.03 \times 10^9 \, W^{0.92} \text{ grams}$$

for the mass thrown out, and assume 7.5% of this comes in contact with radioactivity. This 7.5% is assumed to have a log-normal mass distribution with 50% of the mass in particles of less than 100 μ diameter and 0.4% of the mass in particles larger than 10,000 μ . The fraction of a particle melted will be, for the present, considered independent of particle size.

There is next established a 69th concentric figure, a sphere, which is to represent the vapor phase of the cloud. At the end of the Anderson's first time interval (1 sec.) it is determined which discs have left the cloud. From the yields and half-lives of the primary fission products the quantities of the various product nuclides present at the midpoint of the time interval (0.5 secs.) are calculated. The temperature at this time is also determined. The temperature is used to calculate the vapor pressure of each fission product oxide at a partial pressure of oxygen of 0.2 atmospheres. This is used in conjunction with calculated cloud volume and number of moles of vaporized soil material to determine the distribution of each element between the vapor and condensed states.

The quantities condensed which leave the cloud can now be considered to be initial quantities for a Hunter-Ballou type calculation.[8] The uncondensed radionuclides are considered to be equally distributed throughout the fireball. Again, the Hunter-Ballou type calculation is employed using these uncondensed quantities as starting values in order to compute the quantities present at the midpoint of the next time interval. This process of gradual depletion of particles and radionuclides in the cloud is carried out until the temperature falls to 1400°C.

The second phase of condensation now begins. The portions of each radionuclide present in the condensed phase are now considered to be frozen in. The nuclides in the vapor phase are considered to be either one of two kinds: volatile or involatile. The choice depends upon whether or not their concentration in the cloud is greater than the vapor pressure of the pure substance at the given temperature. At the midpoint of the next time interval the temperature and vapor pressures are calculated as before. Those elemental forms which exceed their vapor pressures are distributed among the particles present according to the surface offered by the particles. Particles leaving the cloud therefore now scavenge radioactivity in proportion to their area rather than their volume and the quantity of each radionuclide remaining behind is equal to that required to saturate the cloud at the given temperature. The process is carried out until all discs have left the cloud.

It is now seen that the rate of arrival of any given radionuclide at any point can be calculated as well as the radionuclide distribution at any point at any time after burst. This radionuclide distribution can now be used as the starting point for the calculation of dose rate according to the decay schemes of the radionuclides involved.

Problems of throwout, stem treatment, and environment induced activities are still under consideration.

The calculations described are very complex and the amount of computer time required to make them has not yet been determined. It is not beyond the realm of possibility that the time required for a complete calculation would be prohibitive were it not for certain simplifications that might be made. First of all, there are those substances which are refractory to an equal or greater extent than the vaporized soil. Those mass chains which consist entirely of such elements will be completely condensed from the outset. At the other extreme are mass chains which will be present primarily as gaseous materials throughout the better part of the condensation process. It has been pointed out[1] that the relative abundances of one such refractorily-behaving radionuclide and one such volatilely-behaving radionuclide at any point in the fallout pattern would determine the relative abundances of the other radionuclides to an extent which was quite adequate for the purposes of estimating fractionation and furthermore was grounded more solidly in experimental observation than is the preceding treatment. In principle, therefore, an ability to predict the abundance of two such nuclides, coupled with a knowledge

of the fractionation systematics obtained through observation, would yield a method of accounting for fractionation in fallout which is at once more simple and more reliable than the detailed calculations described. We will return to this subject in subsequent sections.

Part IV. Means of Testing the Miller-Anderson Fractionated-Fallout Model

The model which we have described is a complex combination of mechanical, thermodynamical, physical-chemical and nuclear-physical considerations. The intricate structure rests shakily on a large number of simplifying assumptions and approximations. The questions we now ask are, "How much closer are we now to reality than we were before in our ability to predict fallout patterns?", "What can we do to improve this approach to reality?" and "How much of this do we want to do?" The first two questions will be discussed in these last two parts of this paper.

One thing we hope to do very soon is to apply the considerations described here to the treatment of data from Operation JANGLE shots. When we do we may achieve better agreement between predicted and observed values than have been obtained before or we may not. Improved agreement would naturally be more encouraging. But how could we be sure that this was not obtained by a fortuitous concellation of errors for the particular event studied? And how could we be sure that in some subsequent application these errors would not cancel and we would not be so fortunate? Observational data in the form of dose patterns and decay curves are themselves complex results of the radioactive properties of the radionuclides present in the fallout material. The calculation of these quantities from the radionuclide composition is a fairly straightforward problem. The reverse calculation is, however, virtually impossible, for while a certain radionuclide composition will result in only one dose and one decay curve, the particular dose or decay curve can be described by a number of radiochemical compositions. It is therefore seen that the radiochemical composition of the fallout at particular points is the basic knowledge that is wanted. It is this knowledge which we expect our fallout model to predict and it is observation of this composition which constitutes one of the most desirable types of data for determining the reliability of the predictions. The quality of radiochemical data available for Operation JANGLE shots is very poor by present day standards and so our uncertainty will be increased by having to rely upon the less desirable and more complex type of observation.

Land surface shots in the Eniwetok Proving Grounds are better sources of material for testing fractionated fallout predictions. Shots Zuni (1956) and Koa (1958) in particular, were the subjects of considerable useful documentation. However, the chemical and thermal properties of coral differ so greatly from those of Nevada Test Site soil and other surfaces to be expected within the continental limits of the United States that success in these areas would be no guarantee of success in the latter cases. Nevertheless, the application of our fallout model to these

bursts appears to be a worthwhile method of testing and one which we intend to do.

The assumptions involved in Miller's approach can be thought of as falling into two groups. First, there are the assumptions bearing upon the distribution of radionuclides between vapor and condensed phase in the first stage of condensation. These are, namely, thermodynamic equilibrium and applicability of Raoult's Law. The second group of assumptions are those which are required to estimate the amount of molten soil present in the fireball at the time of condensation. In view of the tenuous nature of both of these sets of assumptions and desirability of testing each set independently of the other should be evident. To test the applicability of Raoult's Law and thermodynamic equilibrium independently requires a reliable knowledge of: (1) the quantity of molten soil; (2) the existence of a single vapor-liquid phase separation; (3) the time of separation; (4) the temperature at this time; (4) the volume of the vapor phase at this time; and (6) the resulting distribution of fractionated nuclides. This combination of requirements appears to be nearly fulfilled in the case of the Rainier shot. The Rainier shot occurred at the Nevada Test Site on September 19, 1957 about 800 feet underground with a yield of 1.7 KT. It appears to have vaporized 1.5 tons of rhyolite and melted another 850 tons. This molten soil was in contact with the vapor phase for a period of about 30 seconds to 2 minutes after the time of burst. At this time there appears to have occurred a sudden drop in pressure due to the venting of the vapor phase into some external volumes. The temperature at this time was not uniform throughout the system but there is evidence that condensation had already occurred in the vapor phase. The temperature in the vapor phase was therefore less than 2200°C. and the temperature of the lowest or coolest molten portion was greater than 1400°C. The volume of the vapor phase at the time of separation is known from the measured dimensions of the cavity. Fractionation data of good reliability is available for a significant number of radionuclides at several representative locations. It is from these data that the estimate of the time of separation has been made. This time is long enough so that much of the uncertainty in independent yields and half-lives of short-lived fission product will not significantly influence the calculations. The quantity of moisture present in this burst was much greater than would be expected in a Nevada surface burst. The effect of this may be neglected as the first approximation. The most serious drawback appears to be the difference in subdivision of the molten material in this case compared to that in a surface burst. The assumption of thermodynamic equilibrium includes an assumption that the radionuclides in the condensed phase have had the opportunity of distributing themselves uniformly throughout the molten volume. The speed at which this is accomplished depends upon the diffusion coefficient of the substance in the molten material. The diffusion constants of substances in silicates can be 10 or 100 times lower than the diffusion constants of other high temperature materials such as, for example, molten salts. Glass manufacturers are potential sources of additional information.

If the material under consideration is in a fine state of subdivision this will be less important. In the Rainier shot the material was essentially undivided. In order to have a reasonable hope of success it therefore would appear that this should be accounted for in the calculations involved in the model before it is applied to Rainier data. Needless to say, such a refinement would materially improve the realism of the approach taken in the initial model also. Therefore this is something which appears to be worth doing as part of the model refinement process.

Two kinds of experimental measurements which we are planning to carry out in the laboratory will help to alleviate our ignorance in these matters. First, we will measure the rate at which various fission products diffuse through molten silicates and determine if the rates are fast enough to permit equilibrium. Secondly, we will determine the ideality of behavior of various fission product oxides over molten Nevada soil. Both these types of experiments will not only yield evidence of the validity of the assumptions involved but will also yield information by which the assumptions can be corrected and more valid treatments incorporated.

Part V. Fallout Data Required for Testing

In this section I want to outline the kinds of observation which would be more or less ideal for testing the type of model that we have described and obtaining additional data which we need. These thoughts are put forth with little regard for the economics involved. In some cases order of magnitude estimates of the costs can be given. The decision as to whether a foundation for fractionated fallout models of the firmness to be obtained by basing it on the data described is worth the price of investigation is beyond the scope of these studies.

The main areas of ignorance have been summarized by Knapp.[9] This summary, which I now quote, forms the basis of this part of my presentation.

> "What we need to know, and what we generally do not know to within a factor of 2 to 3, may be partially described in the following way.
>
> (1) The distribution of the radioactivity created in nuclear explosions between stratospheric, tropospheric, and local fallout. This distribution depends strongly on the location and conditions of burst, and on the yield of the weapon.
>
> (2) The size distribution, physical and chemical form and properties of the fallout particles, and the distribution of fission products and induced activities among them.

(3) The degree and manner in which radioactivity from various types and conditions of bursts is fractionated. Different relative concentrations of the various nuclides may occur in stratospheric, tropospheric, and local fallout than in the total debris, and even within the region of local fallout the relative concentrations may vary significantly with distance from the explosion. Radiological fractionation may also be considered as occurring between particles of different sizes, different chemical form, and perhaps in other ways such as degree of solubility. Such factors need identification, study as to cause, and evaluation as to relative importance."

I can think of no better means of obtaining the answers for the first two areas of ignorance than that used in Project 2.8 of Operation HARDTACK[10] which I will not describe.

The primary objective of this project was to estimate the partition of Sr^{90} and Cs^{137} between local and long-range fallout in nuclear detonations over land and water surfaces. A secondary objective was to determine the spatial distribution of radioactivity and particles in the cloud a few minutes after detonation. These objectives were to be achieved by radiochemical analysis of the following types of samples:

1. Aircraft samples of particles and radioactive gases present in the upper portion of the clouds.

2. Rocket samples of the particulate matter in the clouds to be collected along nearly vertical flight paths, at several distances from the cloud axis.

3. Fallout samples collected along height lines at an altitude of 1,000 feet by aircraft.

Portions of particulate samples were separated into coarse and fine fractions (greater or less than 25 μ, the selected division between local and worldwide fallout) with a Bahco centrifuge and fall rate distributions for the two fractions determined with a micromerograph. These samples were analyzed separately.

The observations were carried out on high yield coral, water, and coral plus water surface bursts. A fractionation correlation of the Sr^{89}, Sr^{90}, Ce^{144} data is shown in Figure 7. Unfortunately, Zr^{95} was not determined in these samples, and Ce^{144} was chosen as the refractorily-behaving standard. The similarity to previously correlated data, from which the correlation line was taken, is obvious. In the land surface burst, it appears that Sr^{89} and Sr^{90} fractionate from each other in the coarse particles, but not in the fine.

Unfortunately, the rocket sampling part of the program failed and the

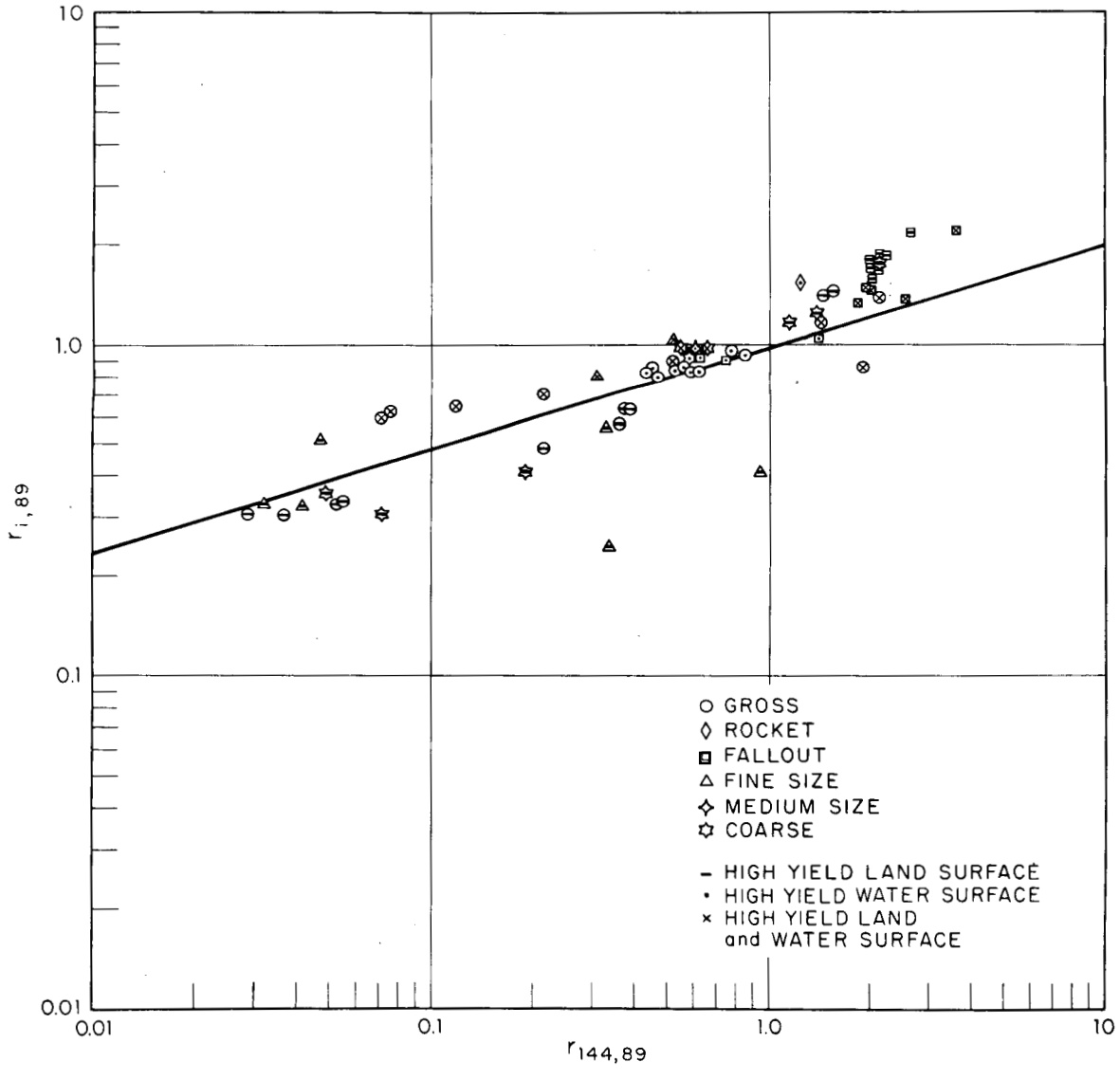


FIGURE 7. FRACTIONATION CORRELATION OF SR^{90}, SR^{89} and CE^{144} DATA FROM HARDTACK PROJECT 2.8 (THE LINE IS TAKEN FROM THE ANALOGOUS CORRELATION IN (1)).

project fell short of its goal. However, much valuable information and experience was obtained, and similar projects in the future would be highly desirable, with even more extensive and intensive documentation, particularly in the study of small particles.

Regarding the third area of ignorance mentioned by Knapp, what we require are fallout patterns which are basically different from the ones usually encountered. The familiar fallout pattern is a series of contours of dose rates at 1 hour after burst. This apparently represents the information most readily interpreted by military and civilian defense planners. It is far too unrevealing to allow adequate building and testing of a fractionated fallout model. What we need in any given burst is not one set of fallout contours, but two. The first set of contours would show the fraction of some refractorily-behaving radionuclides per unit area. For example, one might show the fraction of the total amount of Zr^{95} produced by the device which can be found per square foot at the locations indicated. The second set of contours might represent the same type of information for some volatilely-behaving radionuclides such as Sr^{89} or Cs^{137}. This would provide part of the required information but it would be more instructive to have for the second set of contours a chart of the ratio of Zr^{95} to Sr^{89}. We have called the logarithm of this ratio the "fractionation index." Fractionation index contours would clearly show how the degree of fractionation varied throughout the fallout pattern.

We would now be in possession of information concerning an extensive parameter to indicate the quantity of debris at a given location and an intensive parameter to indicate the departure from representativity at any location. We would still need data to be able to fill in the distribution of the other fission product radionuclides and important induced activities. If we were trying to predict the distribution of each nuclide with our model we would need these data in order to see if we had predicted the distribution properly. On the other hand, if we were only using our model to predict the Zr^{95} contours and the fractionation index contours, we would need these data to interpolate empirically the fractionation of the other radionuclides. Presumably, these data would require an exhaustive radiochemical analysis of a limited number of samples selected for their spread in fractionation index.

Such data could also be used to integrate the fractions of various radionuclides which deposited. By mass balance considerations, the amounts contributed to worldwide fallout could be calculated. Consistency with the data described above would offer valuable support to the results. Experience with field operations has led to the conclusion that whatever can go wrong, will go wrong, and planned over-documentation can turn out to be inadequate documentation.

One would of course want to obtain much other information at the same time such as dose rates, decay curves, gamma assays, and gamma spectra,

mass of fallout, of particle size data, individual particle analysis, etc., which are usually collected during a field test. If this kind of information were backed up by the kind of documentation I have described it is easy to visualize how the correlation of this knowledge with basic radiochemical information would permit a more meaningful interpretation and applicability to prediction than has ever been realized in the past.

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PARTICLE FORMATION AND FRACTIONATION IN AIR BURSTS

Edward C. Freiling
U. S. Naval Radiological Defense Laboratory
San Francisco, California

ABSTRACT

The relation of fallout formation processes in air bursts to processes occurring in other types of bursts are discussed. Results obtained to date on testing available particle formation theories and means of extending these theories to include fractionation are described. A brief summary of available results of fractionation correlations on air burst debris is given and data requirements from future detonations are presented.

Introduction

In this paper I will describe the long-range portion of the fallout model program at NRDL. The object of this portion of the program is two-fold: first, to refine the treatment of land surface bursts described in the previous paper; and second, to extend model-making capability to other types of bursts in line with the general objective of the program. The applicability of the study of air bursts to the latter objective is evident. The applicability to the first objective will be discussed in Part I below on the importance of air bursts. Part II will discuss gross phenomenonolgy and Part III will discuss the macroscopic features of the formation of air burst debris. Part IV will treat the interaction between the macroscopic debris and the product radionuclides and Part V will point out data required from future bursts.

In the remainder of this paper we will talk as much of plans as of

results. Nevertheless, even the preliminary state of accomplishment which we have reached has yielded information of interest and value.

Part I. Importance of Air Bursts

The knowledge obtained from the study of air bursts is applicable to the study of land surface, water surface, and tower bursts. The reason for this can be seen by considering the processes involved.

Fig. 1 outlines the processes which occur in a land surface burst. It is a complex picture of simultaneously occurring nuclear, physical and chemical reactions. The nuclear processes are shown at the top of the figure, namely, the formation of the primary induced and fission product activities followed by their decay into daughter activities or into stable nuclides. Below these are shown the physical-chemical processes, i.e., environment vaporization followed by nucleation in the cooling cloud, condensation, and scavenging by late-arriving particles. The chart is divided timewise on the horizontal scale into a blast phase, and a fallout phase. The chart is a gross simplification but it serves to orient the reader to the interplay of the various features involved in the overall picture of fallout formation.

With regard to the nuclear processes, although there is much missing data concerning independent fission yields and the half-lives of short-lived fission products, the means of estimating the missing parameters have been the subject of considerable study. The methods of estimating the unknown quantities are therefore relatively straightforward, if not completely satisfactory. Certainly we are on much firmer ground in this part of the picture than in the lower part. Before considering the interaction of the radionuclides with molten and solid particles in various stages of transition it is necessary to have some understanding of particle formation itself. Chronologically the first process to consider is that on nucleation. This if followed closely, and in part simultaneously, first by condensation and then by agglomeration. Nucleation is the first step in the transition from a vapor to a condensed phase, whereby clusters of condensing atoms or molecules finally attain a thermodynamically stable condition of existence. While nucleation is occurring in one part of the cloud, nuclei already formed in another part will be condensing vaporized material on their surface. As the number and size of the particles becomes sufficiently large the possibility of particles colliding and coalescing becomes greater, and at the same time, due to depletion of the carrier material in the vapor phase, the nucleation rate bcomes less.

The particles formed by these processes will be in the small size range, probably of the order of 20μ or less. When the temperature of the cloud has fallen to the point where incoming soil particles are no longer vaporized, these new particles will scavenge the previous ones. The

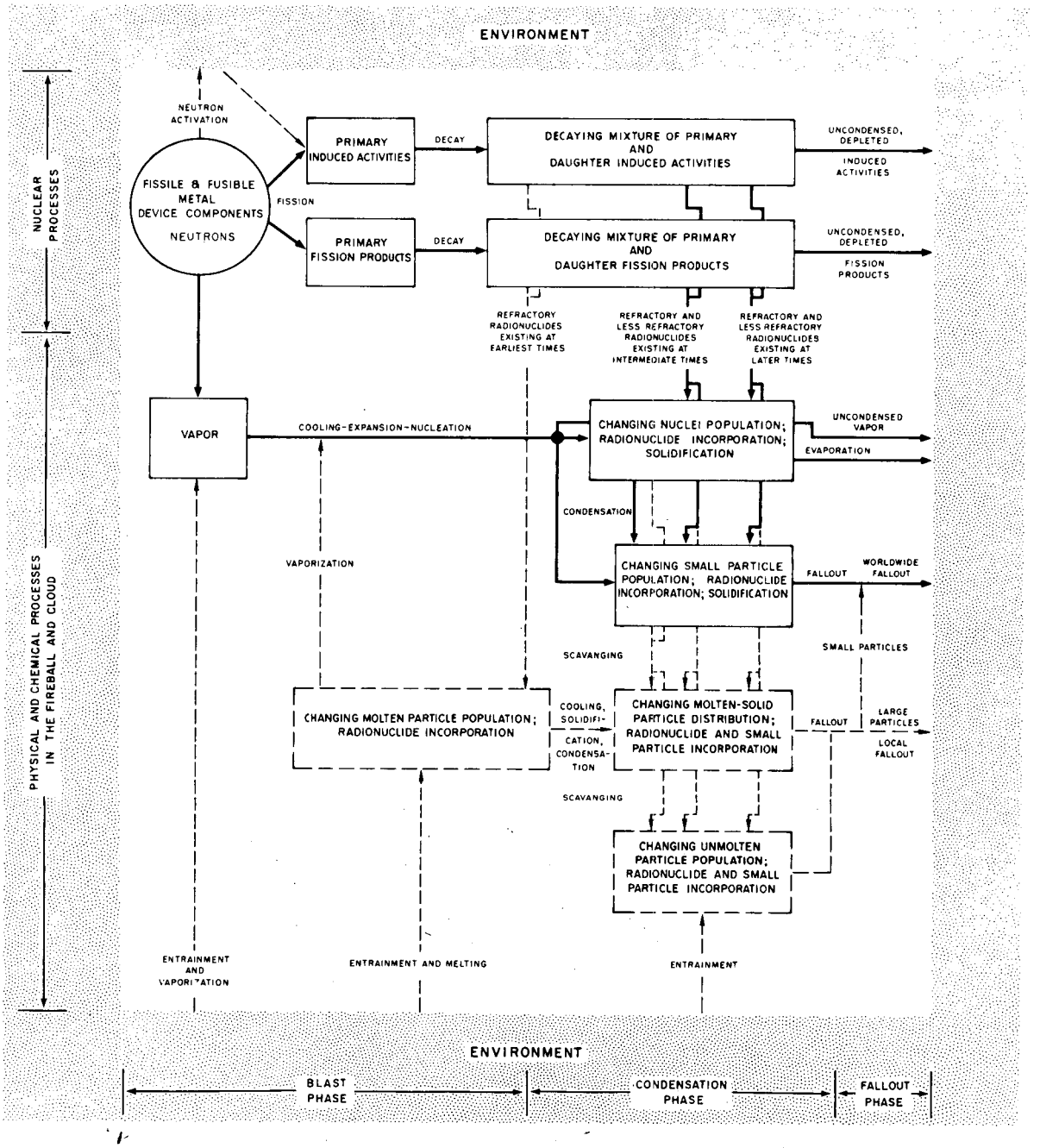

FIGURE 1. Fallout Formation Processes in Air and Surface Bursts.

number remaining unscavenged, together with the small incoming particles, will constitute the portion of the debris contributed to tropospheric and stratospheric fallout (cf. Parts (1) and (2) of the requirements outlined by Knapp and quoted in the previous paper, page 42).

Now it can be seen that subsequent interaction of these primary particles with engulfed but unvaporized soil material will obscure the effects of the primary processes that one sees by examination of the debris. In order to test a theoretical treatment of nucleation and condensation by the properties of such debris, one would have great difficulty not only in sorting out that portion of the observed data due to the processes of interest, but also in dealing with large uncertainties in the quantities of soil involved. On the other hand, in an air burst, such a theory could be tested with much greater hope of success for two reasons: first, the quantity of material vaporized is accurately known; and second, the processes are not complicated by scavenging.

There are other features of air bursts which make them important in themselves. First, whereas land surface bursts contribute only a fraction of their radioactivity to world-wide fallout, air bursts contribute virtually 100% of their activity to world-wide fallout. Secondly, air bursts are valuable sources of data for the prediction of fresh water surface burst phenomena such as might occur in the Great Lakes. Here the initial debris formation processes should be very similar to those occurring in an air burst, the presence of vaporized water being neglected as a first approximation. Subsequent interaction between the initial particles and the condensed water could be elucidated by laboratory studies of the interaction of air burst debris with water at high temperatures. The application of seawater surface burst data to the prediction of fresh water surface burst data is not as reliable as might be thought at first sight, due to the presence of large quantities of salts from the vaporized ocean water and the fact that most seawater surface bursts have been barge shots and have not covered a very wide yield range. The extent to which the presence of barge material and salts has altered the fundamental fallout particle formation processes is not known.

Finally, as will be seen from the subsequent part of this paper, there is a good possibility of reaching a fairly complete understanding of the subject of air bursts at an early date. Tower shots would then form a link between an air burst model and a land surface burst model because the initial particle formation processes in a tower shot would be dominated by the presence of iron, as are air bursts, but would have the additional complication of soil, as in the case of surface bursts. (See Fig. 4 of the preceding paper, page 33).

Part II. Phenomenonology

An air burst begins at a time called the zero time, t_o, with the release of a large amount of energy W in a small space called the fireball. The partition of energy between thermal and mechanical effects is different for "high altitude" bursts (defined as bursts at heights above 100,000 ft.) than for air bursts at moderate heights. We will limit our discussion to bursts at moderate heights. Then about 5% of the energy released is in the form of initial nuclear radiation (gamma rays and neutrons) and 10% in residual radiation (alpha, beta and gamma rays). The remainder is rather evenly dividied between mechanical energy (blast) and thermal energy (heat and light). The following description of subsequent phenomena is a blend of those found in "The Effects of Nuclear Weapons" and in Hillendahl's work.(1)

To quote The Effects of Nuclear Weapons: "Because of the very high pressure within the exploding bomb the residue, consisting of fission products and all other bomb materials, moves outward from the center of the explosion at a very high velocity... After a few microseconds, nearly all of the debris is contained in a relatively thin shell of high density called the 'hydrodynamic front': its initial temperature is about a million degrees and it is traveling at a speed of several hundred miles per second. ....The mass energy is transferred to the medium" and a blast front is formed. At first this is preceded by a radiation front (the surface of the fireball)," because the mean free path of the radiation in the hot gas is so long that the transfer of energy by radiation is more rapid than by mass motion." As the temperature drops, the transfer of energy by radiation becomes less rapid, the shock front begins to advance more rapidly and passes it at a time when the temperature has fallen to about 300,000°C. This phenomenon is called the "hydrodynamic separation." It occurs at about $10^{-4}\ W^{0.42}$ sec. at a radius of about $4W^{0.37}$ m. After this the fireball consists of two concentric regions. The inner one is called the isothermal sphere and is bounded by the radiation front. Its radius is R_2. The outer region consists of luminous, shock front heated air. It is bounded by the shock front, whose radious is R_4. (See Fig. 2 of previous paper, Page 30). The radius then grows accoring to the law

$$Wt^2 = 1.27 \times 10^{-8} \rho_o (2R_4)^5$$

where R_4 is in meters, t the time in milliseconds, and ρ_o the ambient air density in g/l. The luminous air produced by the shock front obscures the vision of the isothermal sphere until a time of $3.65 \times 10^{-3} W^{0.42}$ sec. called the "breakaway." At this time the apparent temperature is a minimum and the isothermal sphere again becomes visible. The temperature then rises again to a maximum value of $8900\ W^{-0.03}$ degrees K at a time of $0.045\ W^{0.42}$ sec. This time is called "the time of final maximum" and given the symbol t_f. The temperature drops to $3300\ W^{-0.03}$ degrees K at a time of 15 t_f. Its logarithmic dropping rate in this period is $(t/t_f)^{1/40}$. At this time (15 t_f) the

radius of the isothermal sphere has levelled off to a value somewhat exceeding 63 $W^{0.35}$ m. The radiated power equation is given by

$$H(W,t^*) = 6.68W^{0.58}\, t^{*-1.6}{}_e{}^{-9e^{-2.73t^*}}\, e^{-9e^{-1200t^*}}$$

according to which 0.55W eventually becomes radiated. Here $t^* = t/t_f$.

The cloud during this time has been rising. For a 1 MT burst the rise in feet is given approximately by $76{,}000\ (1 - e^{-0.42t(\text{sec.})})$ out to about 10 min.

Part III. Particle Formation

Three theories of particle formation have been found which apply to air bursts. These consider the basic processes previously mentioned in various detail and lead to particle size distributions in terms of two basic parameters: the total number of particles and a characteristic particle size. The shape or functional form of the distribution depends upon the relative importance given to the processes treated. The characteristic size depends upon various measurable or estimable parameters. Once these have been fixed, the total number of particles is determined by the mass of the device and the demand of mass balance. It is expected that the details of particle formation mechanisms will be most accurately revealed by the theory which best fits the observations, and therefore that theory would form the starting point for developing additional applications.

The first theory, due to Woodcock (2), will be quickly dispensed with. First, because it is described in a confidential report, and second, because it has not been found to fit the observed data. The treatment could concievably be more applicable to other burst conditions.

The second is due to Magee.(3) His treatment emphasizes the nucleation and condensation processes and arrives at the exponential distribution

$$n(r)\,dr = \frac{n_T}{\bar{r}} \exp\left(-\frac{r}{\bar{r}}\right) dr$$

where n_T is the total number of particles and $\bar{r}$ is the mean radius. In this theorty $\bar{r}$ is the ratio of β, the radial growth rate of a particle, to α, the logarithmic growth rate of the nucleation rate I.

The third treatment was devised by Stewart (4). It considers the process of nucleation, condensation and coagulation occurring simultaneously. Stewart arrives at the log normal distribution

$$n(r)\, d(\ln r) = \frac{n_T}{\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\ln \frac{r}{\bar{r}}\right)^2\right] d(\ln r)$$

Here, again, n_T is the total number of particles. The modal radius is given by

$$\bar{r} = \frac{v_B N_o T_o}{Kn} \left(\frac{k}{2\pi mA}\right)^{\frac{1}{2}}$$

where

v_B = molecular volume in the liquid phase

N_o = initial number of atoms per unit volume

T_o = absolute temperature at the time of condensation

$K = 4\ kT/9\eta \approx 3 \times 10^{-9}$, T = absolute temperature, η = air viscosity

n = concentration of nuclei

k = Boltzmann's constant

m = molecular mass

A = 7500°K.

As mentioned above, the most fundamental property of the predicted distribution is their functional form, or what is equivalent, their shape. These predictions can be compared with observed data in various ways: by using either differential or integral curves and by comparing either slopes, areas or individual values. All of these means were considered. The method found to be most suitable, not only for determining the shape, but also for evaluating the parameters, is based upon converting these equations to linear form. Thus Magee's equation

$$n(r) = \frac{n_T}{\bar{r}} \exp\left(-\frac{r}{\bar{r}}\right)$$

becomes

$$\ln n(r) = \ln \frac{n_T}{\bar{r}} - \frac{r}{\bar{r}},$$

so that a plot of ln n(r) vs. r will have slope - $1/\bar{r}$ and intercept $\ln \frac{n_T}{\bar{r}}$, similarly, Stewart's equation

$$n(r) = \frac{n_T}{\sqrt{2\pi}} \exp\left[-\frac{1}{2} (\ln r/\underline{r})^2 \right]$$

becomes

$$\left[2 \ln n(r) + (\ln r)^2 \right] = \left[2 \ln \frac{n_T}{\sqrt{2\pi}} - (\ln \underline{r})^2 \right] + 2 \ln \underline{r} \ln r$$

so that a plot of $\left[2 \ln n(r) + (\ln r)^2 \right]$ vs. ln r will have a slope of 2 ln $\underline{r}$ and an intercept $\left[2 \ln \frac{n_T}{\sqrt{2\pi}} - (\ln \underline{r})^2 \right]$.

This technique has been applied to particle size data from a number of air bursts. It has resulted in discarding Woodcock's treatment in favor of the other two, but has not permitted a choice to be made between the treatments of Stewart and Magee. An example of its application in the latter two instances is shown in Figs. 2 and 3. The scatter at the large particle end of the curve is not statistically significant, due to the small number of particles involved. The value of 5μ is taken as the lower limit of reliability of the data. The values for the parameters obtained are: $n_T = 1.86 \times 10^6$, $\underline{r} = 0.0718\mu$; Magee, $n_T = 2195$, $\bar{r} = 1.83\mu$.

From these results one can calculate the following properties of the sample:

	Stewart	Magee
Total Surface	0.89 mm^2	0.185 mm^2
Total Volume	$2.65 \times 10^5 \mu^3$	$3.38 \times 10^5 \mu^3$
Total weight (assuming density 2.5)	652 smidgins	845 smidgins

From these data it is seen that particle size separations would have to be done in addition to chemical analysis in order to obtain the information required to eliminate one of these theories. If the surviving one were also able to predict the characteristic particle size with reasonable accuracy, one could proceed to apply the treatment with some degree of confidence that the processes involved were well understood and properly treated.

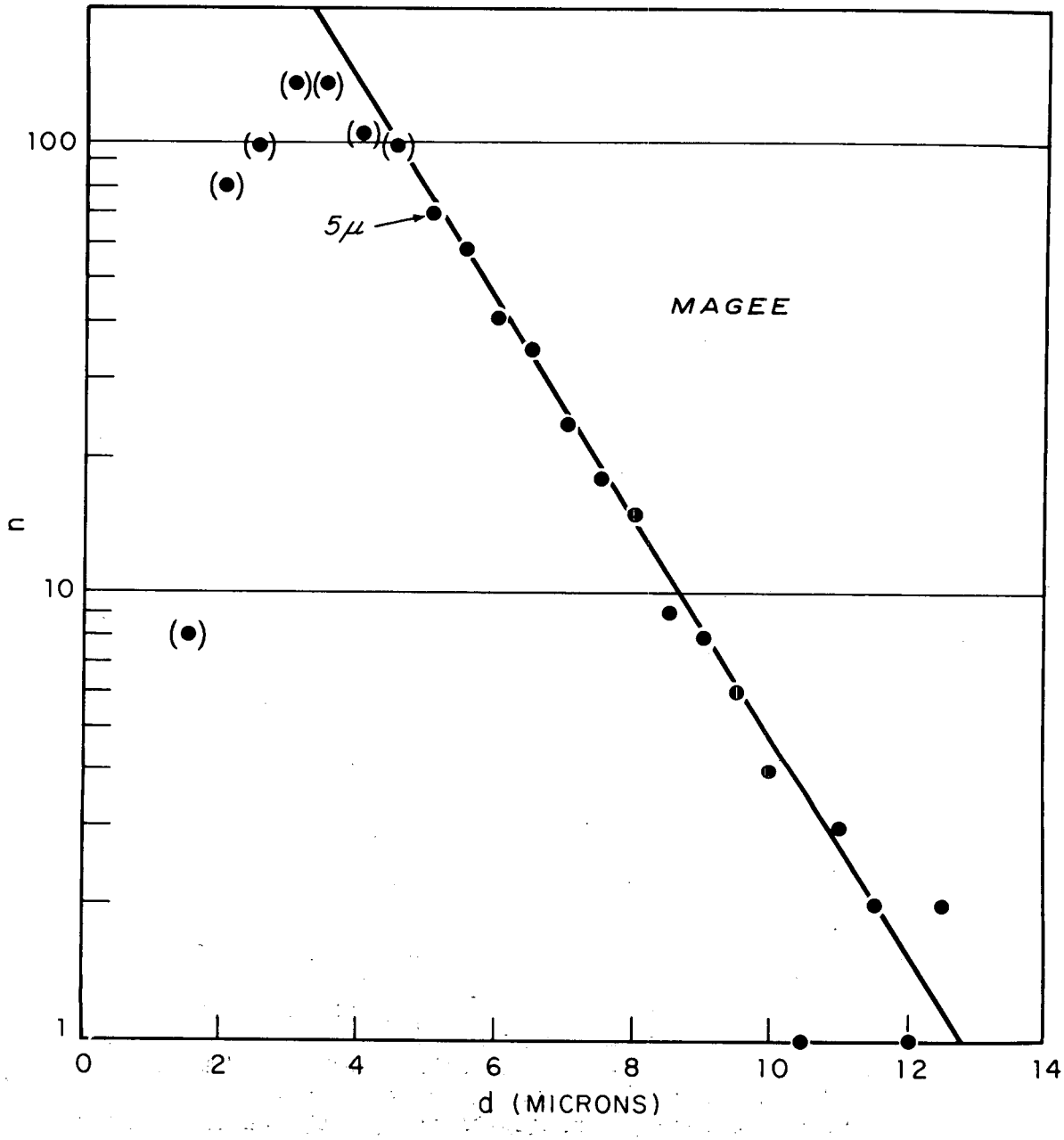

FIGURE 2. PARTICLE SIZE DATA FROM AIRBURST DEBRIS PLOTTED ACCORDING TO MAGEE'S THEORY. (DATA BELOW 5μ ARE BIASED).

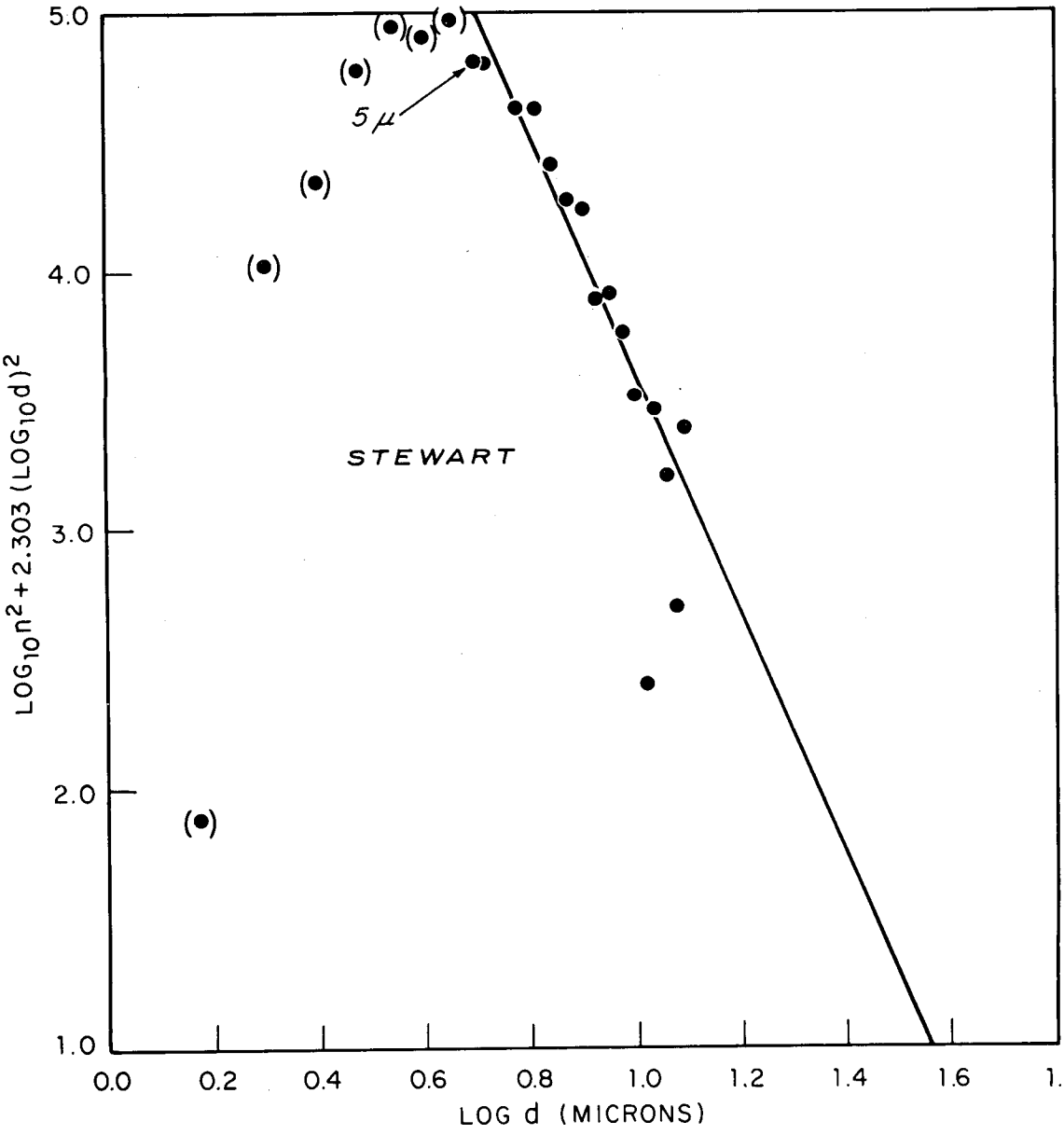

FIGURE 3. THE DATA USED IN FIGURE 2 ARE HERE PLOTTED ACCORDING TO STEWART'S THEORY.

Part IV. Radionuclide Incorporation

As indicated previously, the fractionation resulting from the interaction of radionuclides with the nascent particles can also be thought of in two regards: first, the fractionation index, or ratio of two extremely different behaving product radionuclides; and second, the interpolated behavior of the other products. The fractionation index is the result of such vastly different properties of the mass chains involved that there is hope that it can be treated by a very simple first approximation. This is, that the refractorily-behaving nuclide will be distributed among the particles according to their volume or the cube of their radius while the volatile radionuclide will be distributed according to the surface of the particles or the square of their radius. This is a result of the assumptions that (1) the particles are solidified before the volatilely-behaving radionuclide condenses and (2) that the particles remain in intimate contact with the vapor phase during the entire condensation period. From the distribution data for the debris sample in the previous section one calculates that the fractionation index for a given particle size would be log (0.18 r) according to Magee but log (1.1 r) according to Stewart. The former relationships would require 100 μ particles to account for the degree of fractionation observed, if the mechanism described above is realistic. The behavior of some of the intermediately fractionating radionuclides may also be handled fairly well on such a simple basis, while others will require a more refined treatment. This is visualized as involving considerations of collisions between nascent particles and vaporized fission products which lead to incorporations of the products in the carrier material with certain efficiencies (accommodation coefficients) and probabilities of escape. Additional refinements, if necessary, might include consideration of diffusion in the particle, correlations of accommodation coefficient with compound formation, and the extension of condensation theory to account for depletion of source material and for the inhomogeneous development of the fireball.

An important part of the work on the development of an air burst model is the accumulation and correlation of data with which to test it. Data have been gathered from the radiochemical analyses of fractionated samples from a large number of air bursts. The yields of these bursts range over a factor of several hundred. As in the case of high yield surface burst correlations, the logarithm of the ratio of the fraction of total Sr^{89} present in given sample to the fraction of total Zr^{95} was taken as the fractionation index. The values of this ratio range over a factor of about 100. Logarithms of various other ratios of radionuclide fractions were plotted against this index. By machine computation these plots have been fitted with straight lines, the intercepts adjusted to zero, and slopes of cumulative plots calculated. Analysis of the results is still in progress, but some qualitative statements can be made:

1. Many, but not all, of the ratios chosen indicate a gratifying constancy of behavior over the range of conditions involved.

2. The relative volatility of behavior observed bears both similarities to and differences from that exhibited in high yield surface bursts, reflecting the identity of the nuclides and the difference in carrier materials involved.

3. The order of increasingly refractory behavior is as follows: Cs^{137}, Sr^{89}, Sr^{90}, Cs^{136}, Cd^{115}, U and Ag radionuclides, Ba^{140}, Y^{91}, Ce^{141}, Mo^{99}, Pu and rare earth radionuclides, Zr radionuclides.

Part V. Data Required from Future Air Bursts

A representative sample of debris in significant size is the sine qua non for meaningful particle and fractionation data from future air bursts. This demand has been difficult to satisfy in the case of past land surface bursts, but the smaller particles present and recent developments in sampling apparatus make it quite reasonable in the case of air bursts. Furthermore, these samples can be collected in different size fractions. If such a sample were available, chemical analysis or activation analysis would reveal the amount of material in each particle size fraction, and from this the particle size frequency distribution could be calculated. Radiochemical analysis of the particle size fractions would test the hypothesis of volume distribution for refractorily-behaving radionuclides and surface distribution for gaseous-behaving radionuclides. Such an experiment appears feasible and would result in virtually complete documentation for the particle formation process. Information still lacking would be distribution of radionuclides within the small particles as the function of radius. For macroscopic substances this is on the borderline of present capabilities but for radionuclies it is quite beyond what we are able to do at the moment.

Valuable data could be obtained from less intensive tests. The minimum amount of information which would be useful would consist of (1) sufficient particle data to establish n_T and $\underline{r}$ or $\bar{r}$ for the respective theory, and (2) the measurement of some critical extensive property of the debris collected, such as the total mass iron content, number of fissions, etc., which could be used to distinguish between the merits of the distributions proposed.

Experiments such as these, carried out on airbursts, balloon bursts, and the smaller particle size fractions from tower shots where the fireball did not touch the ground, tower shots loaded with soil where the fireball did not touch the ground, and residual clouds from land surface bursts, would forge links for the theoretical chain which

would bind fallout formation processes under these diverse burst conditions into a unified structure.

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GAMMA-RAY DOSE FROM SHORT-LIVED FISSION PRODUCTS

Phillip F. Gustafson and S. S. Brar
Argonne National Laboratory
Argonne, Illinois

The external γ-radiation dose due to the deposition of short-lived fission products from weapons tests was largely ignored until the spring of 1959, when several observers noted increases in γ-dose rates attributable to short-lived activity (1,2,3). These relatively high levels of radioactivity seen during early 1959 were due to three factors: (1) the large number of tests conducted in 1958, (2) the short stratospheric residence time of debris from polar detonations, and (3) the spring maximum in fallout deposition. Although a higher rate of deposition during the spring had been noted in previous years, it was attributed primarily to the testing sequence rather than to any meteorological mechanism. As shown in Fig. 1, the spring maximum in Cs^{137} concentration in surface air has been observed annually since 1954. Its occurrence in 1960 and 1961 is strong proof of its meteorological origin. Because the spring maximum coincides in time with higher precipitation, on the average, over a fairly wide latitude band centered around 40-45° North, greater deposition occurs at this time of year throughout the region in question. It is reasonable to suppose that increased deposition, including that of short-lived emitters, also occurred each spring subsequent to 1954, and indeed such has been found to be the case at the Argonne National Laboratory site since 1957.

To the degree that measurements of surface air indicate release of stratospheric debris and hence ground deposition, it is evident from Fig. 1 that the rate of deposition is not constant, even when testing is not in progress. The variation in deposition rate does not imply, however, that there is not an annual rate of depletion of the stratospheric reservoir. Thus one may think in terms of a mean residence time for nuclear debris in the stratosphere. From Fig. 1 it is apparent that a yearly cycle consists of a maximum in spring and a minimum in autumn and winter. By comparing the Cs^{137} concentration in air during successive maxima or minima it is possible to determine a mean stratospheric residence time, as illustrated in Fig. 2 for total Cs^{137} (i.e., Cs^{137} from all tests conducted up to the time of observation).

It has been of interest to examine the variation in residence time as a function of geographic latitude and the altitude of detonation or the altitude to which debris was carried initially. The production of W^{181} in some surface shots and Rh^{102} in a high altitude shot (Orange) of the Operation HARDTACK I series in 1958 have made it possible to assign debris to these two equatorial sources. Activity ratios between appropriate pairs of radionuclides were used to determine the contribution from the Soviet October 1958 polar series. The concentrations of W^{181} and Rh^{102} in surface air at Argonne, corrected for decay back to mid-June and mid-August 1958 respectively, are shown in Fig. 3. Using these concentration figures in conjunction with activity ratios, the total Cs^{137} in surface air was partitioned into that pertaining to HARDTACK I surface and high altitude series as well as Soviet October 1958 series as shown in Fig. 4. The rapidity with which debris from the Soviet October 1958 series was deposited, and the delay in deposition of high altitude debris, materially influenced the amount of short-lived activity reaching the ground from these two sources.

The mean stratospheric residence time for debris from HARDTACK I surface shots as derived from the data in Fig. 4 ranged from 8 to 10 months during 1958-59 to $<$ 15 months in 1959-60. The increase in mean residence time may be explained by the more rapid removal of the low altitude component of this source. The bulk of Soviet October 1958 activity was deposited within one year. The observations of high altitude debris are too limited to determine the mean residence time with any precision, although the continued increase in debris from this source implied that this time must be $<$ 5 years.

The variation in the deposition of short-lived fission products as a function of mean residence time has been studied in some detail by Dunning (4). A similar procedure has been followed here in relating the dose from Ba^{140}-La^{140}, Zr^{95}-Nb^{95}, Ce^{144}-Pr^{144}, Ce^{141}, Ru^{103}, and Ru^{106} to that from Cs^{137}. The ratios for 30-year (genetic dose) and 70-year (lifetime dose) intervals following a test series are shown in Fig. 5 as a function of mean residence time.

By a combination of direct measurement and calculated ground deposition using air and rain concentration data, the ratio of the dose from the aforementioned fission products to that from Cs^{137} was determined at the Argonne site. This was done for debris coming from HARDTACK I surface shots, high altitude, and the Soviet October 1958 series. The ratios for these three sources over 30-and 70-year intervals are indicated in Table 1. The apparent mean residence time for each case, as taken from Fig. 5, is shown in parentheses. The agreement between the residence times interpolated from Fig. 5 and those computed from air concentration measurements is somewhat surprising, particularly when one considers the variability observed in the deposition rate, since a constant rate of deposition is implicit in the ratios plotted in Fig. 5.

The information concerning deposition of debris from the Soviet October 1958 series may be used to draw some conclusions regarding deposition and dose to be expected from the current Soviet tests. The total deposition of Cs^{137} (during 1959) from the Soviet October 1958 series amounted to 40 mc/mi^2 at Argonne. If we assume that this came from 12.5 MT of fission, then 3.2 mc/mi^2/MT of Cs^{137} was deposited within one year. The open-field dose rate during the first year from this concentration of Cs^{137} is approximately 0.1 mr/yr. The integrated 30-and 70-year dose from this level of Cs^{137} deposition as well as that accruing from short-lived activity are shown in Table 2. In order to estimate the open-field dose expected in 1962 as well as the integral dose over subsequent years it is necessary to choose the proper scaling factor. The 1, 30, and 70-year integral doses pertaining to an arbitrary choice of 25 MT as the total fission in the current test series are also shown in Table 2. The average dose arising from natural sources, including cosmic radiation, at the Argonne site is indicated for comparative purposes. Deposition of γ-activity from 25 MT of fission, assuming the same deposition rate and pattern in 1962 as observed in 1959, will result in an open-field dose essentially equal that from natural sources during the first year (i.e. 1962). The open-field dose from this deposition is computed to be 5 and 3% of background for the 30-and 70-year intervals, respectively. A shielding factor of 5 will reduce exposure over 1, 30, and 70-yr. intervals to 20, 1, and 0.5% of background, respectively.

These calculations provide an approximate estimate of exposure due to the current Soviet tests, and may be appreciably altered by the altitude to which debris was carried initially, precipitation, and other meteorological variables. They do, however, establish a procedure for the realistic appraisal of future radiation dose from these tests.

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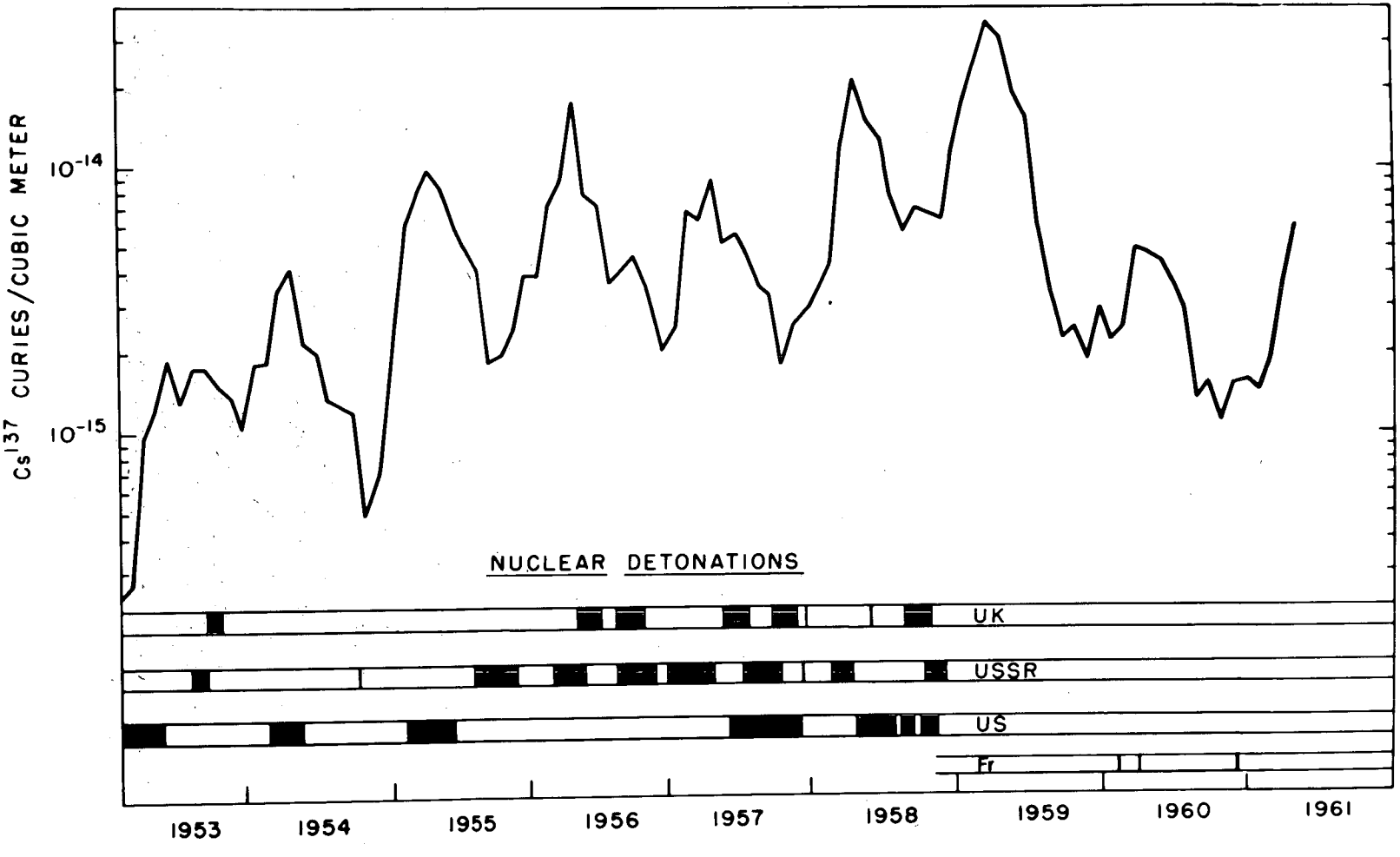

Figure 1. Cs^{137} in Surface Air 1959- 1961

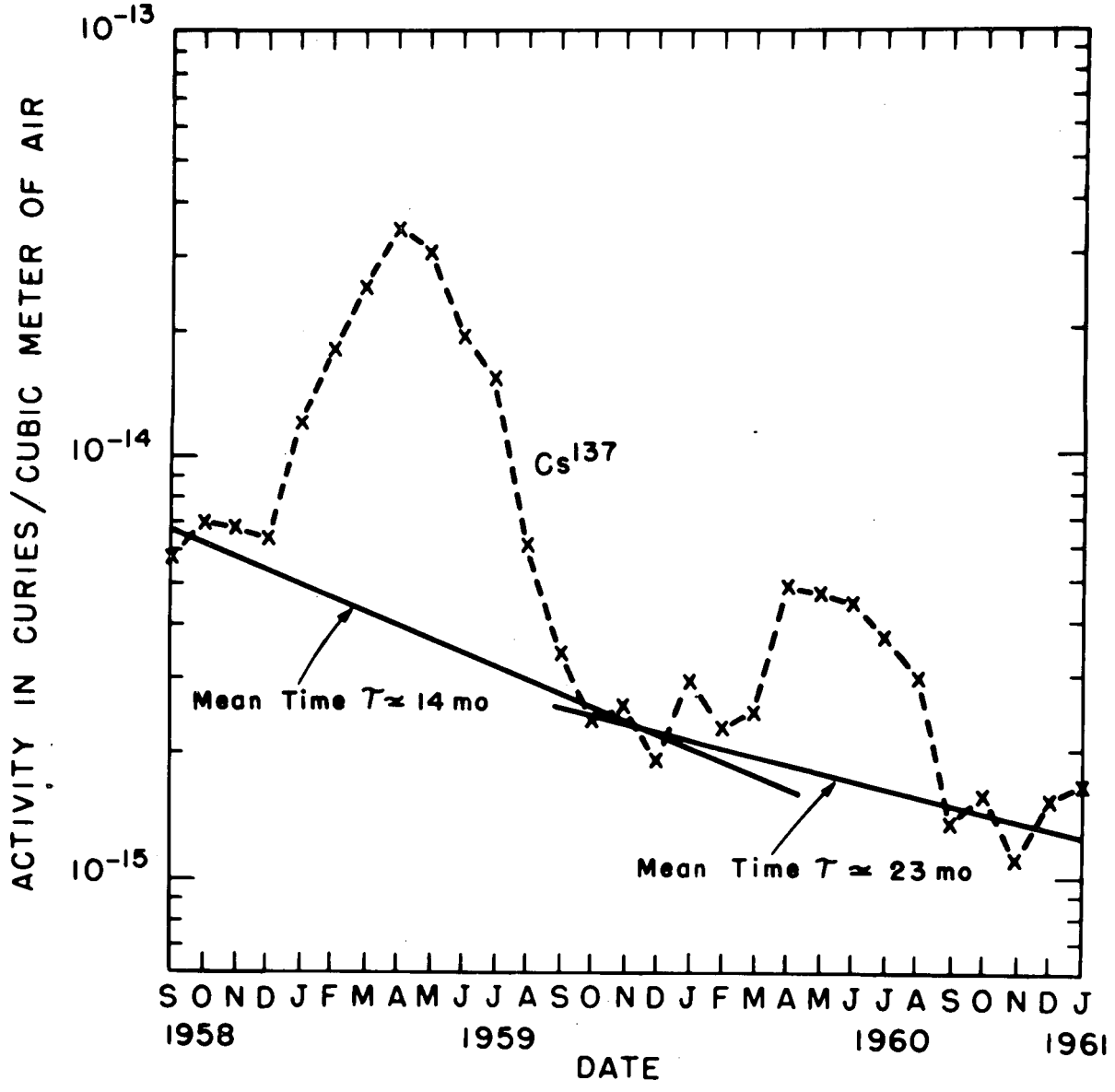

Figure 2. Determination of Mean Stratospheric Residence Time

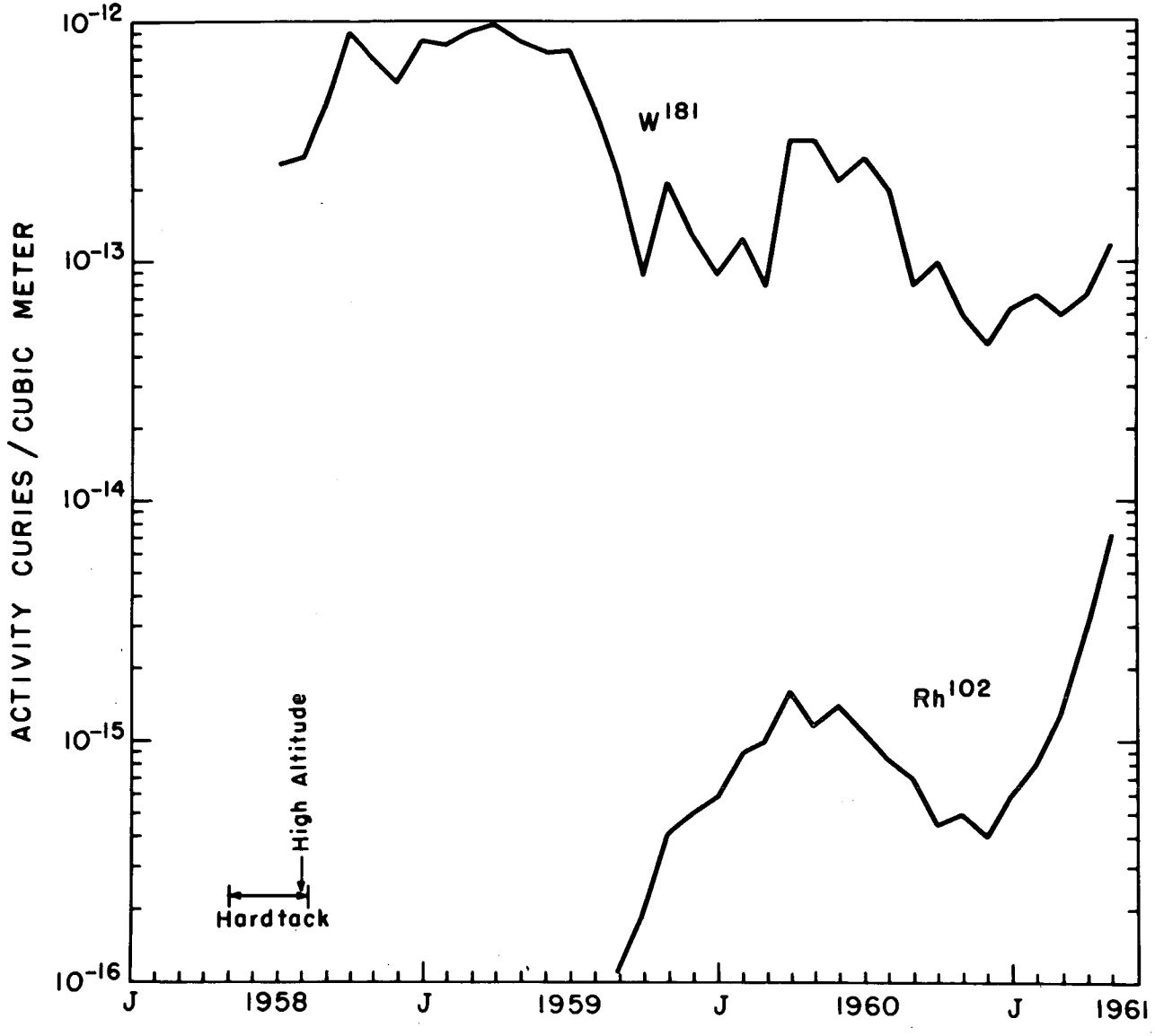

Figure 3. W^{181} and Rh^{102} in Surface Air 1958-1961

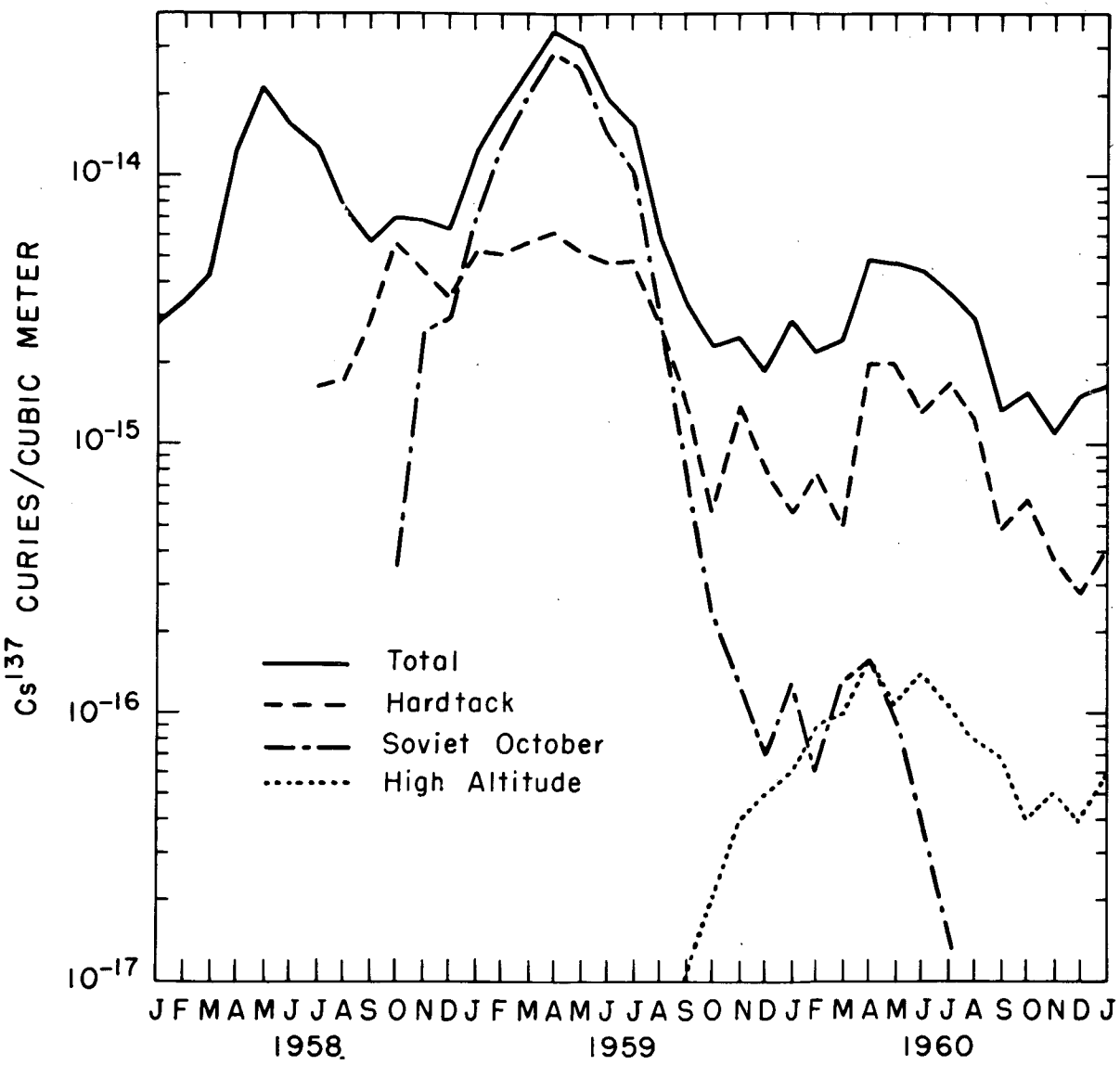

Figure 4. Partition of Cs^{137} in Surface Air

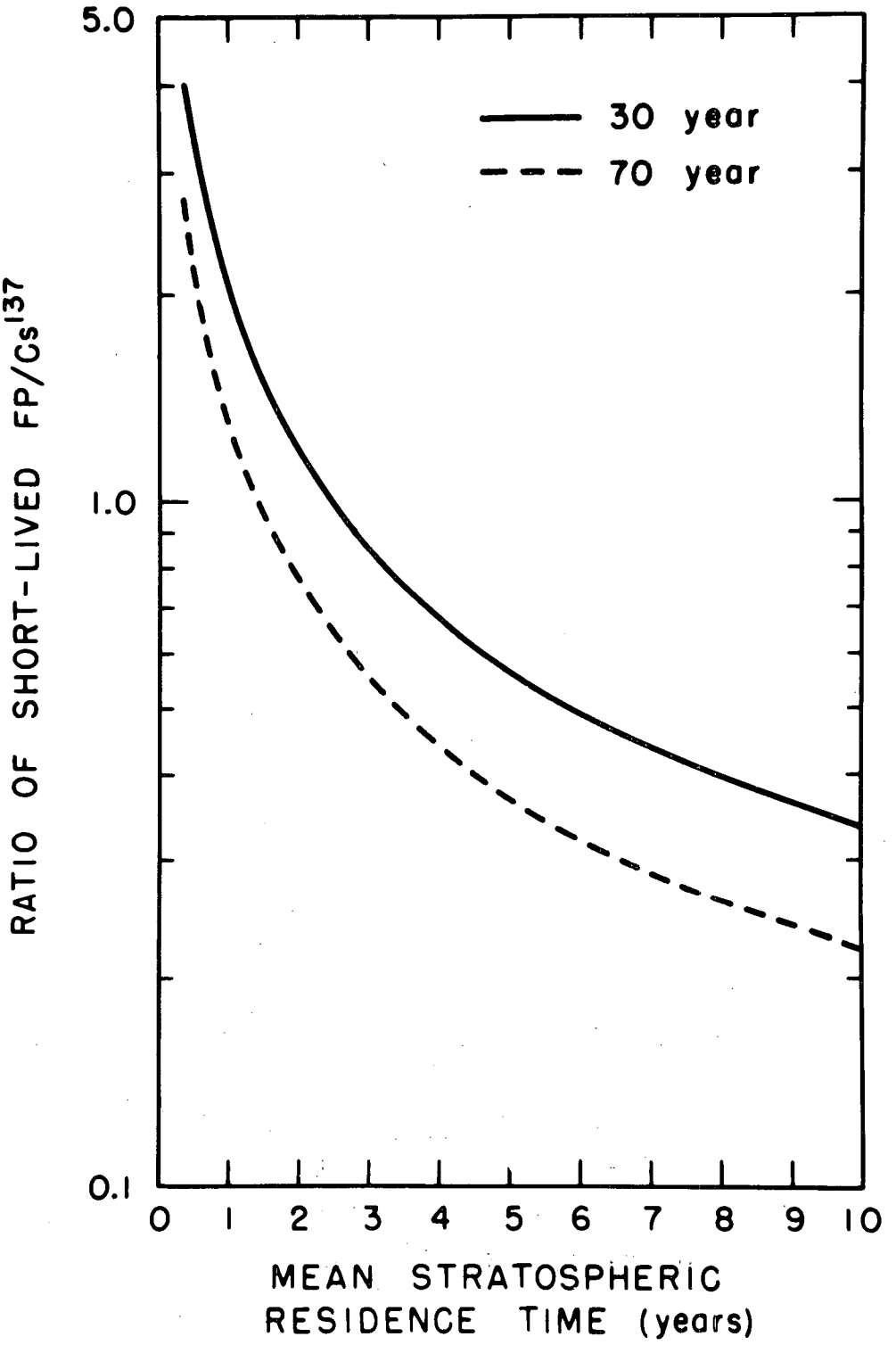

Figure 5. Ratio of Dose from Short-Lived Fission Products to that from Cs^{137} as a function of Mean Residence Time

TABLE 1

Ratio of Dose from Short-Lived Fission Products to that from Cs^{137}

Time Interval	Test Series: Soviet October 1958	HARDTACK Surface	High Altitude
0 30 years	2.21 (8-10 Months)	1.31 (18-20 Months)	0.12 (7-15 years)
0-70 years	1.58	0.90	0.08

TABLE 2

Integral Dose for 1, 30, and 70-Year Intervals
(milliroentgens)

Time Interval	γ-Dose/MT of fission for Soviet October 1958 (observed)		γ-Dose/25 MT of fission for current Soviet series (calculated)		External Dose from natural sources
	Cs^{137}	Short-lived activity	Cs^{137}	Short-lived activity	
0.1 year	0.1	3.83	2.5	96	100
0-30 years	2.08	4.80	52	120	3000
0-70 years	3.22	4.80	81	120	7000

DISCUSSION*

Dr. Harold A. Knapp, Chairman of the session, direct the discussion of the papers of the session which follows.

FREILING: I notice that in one of your (LARSON's) slides, the ratio of Cs^{137} to Sr^{90} decreased as the size of the particles that you were looking at increased. I think this is something that one would expect, but also, as I recall the slide, this ratio decreased for particules which were of the same size as the time of arrival increased. Have you given any thought to the reason for this?

LARSON: No, I have not. We have only recently initiated this particular study. The data presented here are preliminary results.

FREILING: Another question I might ask is whether there are data on any other radionuclides in this debris which were not included in your talk?

LARSON: Yes. We have data on the distribution of Ba^{140}-La^{140}, Sr^{89}, Ru^{103}, ^{106}, Cs^{136} and Cs^{137}, Zr^{95} and Y^{91} in fallout debris. Tissues from animals collected from the same location as the fallout debris was collected were assayed for these samefission products.

HOLLAND: I would like to ask Kermit LARSON whether in his opinion the bone levels (bone Sr^{90} levels in wild animals in thefield, say) from exposure to fallout in the past or currently-occurring fallout is primarily due to inhalation or ingestion, and what relative part you think the inhalation might play?

LARSON: Our data indicates that inhalation is insignificant. What we are dealing with in the native mammals indigenous to a fallout pattern is an ingestion problem. Our studies indicate that the diet, the activity associated with vegetation, is by far the most important factor in the environs of Nevada Test Site (NTS).

* Only the last names of participants are shown after being identified once in the proceedings. Conference speakers are fully identified in the agenda on page iv A list of registered attendees is included on page 236.

Lester VAN MIDDLESWORTH (Univ. of Tenn. School of Med., Memphis): Could the speakers give an acceptable definition of fractionation? There are many different possibilities here. Then, could they suggest a few mechanisms that might account for fractionation?

FREILING: The definition that I use is that fractionation is any alteration of radionuclide composition occurring between the time of detonation and the time of radiochemical analysis which causes the debris sample to be nonrepresentative of the detonation products taken as a whole.

KNAPP: Specifically, how do you measure it with respect to a particular nuclide like I^{131} or Cs^{137}?

FREILING: One can measure any radionuclide from a given detonation and also calculate on theoretical grounds what the yield of this radionuclide should have been in the detonation. By combining the expected yield with the observed quantity one can then calculate back to the number of fissions which were responsible for the sample that one observes. If that sample is unfractionated, the answer is the same for each radionuclide measured. This happens very seldom, however, and usually one gets quite different answers for the different radionuclides that one observes. Maybe we should finish with the definition before going on to the mechanisms.

KNAPP: Is there any base nuclide that you feel worth settling on as being unfractionated by definition, such as Zr^{95} or something like that?

FREILING: We have preferred Zr^{95} because it seems to show a fairly universal refractory type of behavior. By this I mean that it is usually associated with the refractory elements. Furthermore, its fission yield is fairly constant, from one type of formation process to another.

LANGHAM: Kermit (LARSON), you said, with respect to your rodent data, that it was not an inhalation problem, but was associated with ingestion of contaminated vegetation. Would you hazard a guess as to what percentage of this is direct fallout as compared to direct soil uptake from the integral level in the soil?

LARSON: As far as plant uptake of fission products through the root systems of the species of plants that we have been concerned with, we have not been able to demonstrate any root uptake. Therefore, it is my opinion the fission products in fallout material is retained on the leaf surface and in a particulate form. The observations we have made within the areas that we have studied indicate this true.

KNAPP: This is for fairly large particles.

LARSON: Vegetation has been found to be a selective particle size collector within the areas that we study. Based on some 3,000 observations of various species we find the retained particles have a size range of 10 to 14 μ.

Sheldon K. FRIEDLANDER (Johns Hopkins Univ., Baltimore): My question is directed to Dr. FREILING. In the case of air bursts, one can admit that there were probably three general methods for cooling the cloud which is formed after the burst. I can think of radiation and of adiabatic expansion and cooling by mixing of the hot vapor with the cool air. Do you have a preferred mechanism among these, or is there another that you believe that is controlling?

FREILING: Different cooling mechanisms, (or energy escape processes) predominate at different times. About half the initial device energy is lost very quickly as blast energy. Between the breakaway and the time at which a temperature of about 2000°K is reached, the majority of the remaining energy has been lost, and thermal radiation has been the principal cooling process. Its effectiveness decreases rapidly with temperature, however, and below 2000°K the other processes you mention probably predominate.

Irving J. RUSSELL (Air Force Special Weapons Command, Kirtland AFB, N. Mex.): I would like to direct a question to Mr. LARSON. How do you establish the source of your Sr^{90} and Cs^{137} at great distances from the Nevada Test Site (NTS)?

LARSON: We have only determined Sr^{90} and Cs^{137} that does exist out there. If a systematic program were set up to determine if this material might have originated from NTS, I think then we could resolve the question of origin.

KULP: I would like to address a question to Dr. GUSTAFSON. Would you comment on your shielding factor of 5? Does this relate to the open flat field of the farmer, average rolling pasture of forest or to city life?

GUSTAFSON: Yes. The factor of 5 is taken to be that reduction in dose due to one's being outside on a rather flat, even plane, and being inside of structures such as we find in this country that are rather substantial, either brick or frame. It does not take into account any shielding due to uneven terrain or weathering or leaching into the soil.

KNAPP: I would add that this factor is very much unknown, not only what the average amounts to, but what the distribution about the average is. One would hope we would learn more about that quickly, unglamourous as it is.

GUSTAFSON: I would not go along with a factor of two for weathering. That is, some people have frequently used a shielding factor of 10. I believe that this is advocated by the United Nations Scientific Committee. But as far as our soils go in the Argonne vicinity, weathering does not seem to be occurring. That is, if by weathering you mean the downward movement of these gamma emitters, indeed weathering occurs for strontium but perhaps strontium is somewhat unique as far as weathering goes. These other nuclides do not seem to weather, least in this particular soil which is quite high in clay content.

KALKSTEIN: Mr. LARSON, what is the significance of the data on your last graph? It seemed that the bulk of the data would remain on a horizontal line, although a few points were quite high compared to the rest.

LARSON: In 1958, that is, one year after the Smoky shot (Operation PLUMBBOB, 1957) contamination primarily, 41 of the 43 animals had Sr^{90} levels in excess of 10 μμc Sr^{90}/g Ca (strontium units), regardless of weight or age. In contrast, in 1961, 39 of the 53 animals had bone levels less than 10 μμc Sr^{90}/g Ca. The higher levels were restricted to the heavier or older animals. If one accepts the average life of about 3 years for this species of rabbit living in this environment, the 1961 rabbit collection consisted, for the most part, of animals who were not in the direct fallout from this shot nor for the remainder of that season. One suggestion apparent from these data is that an animal has to live in fresh fallout in order to accumulate the higher amounts of Sr^{90}. The environment has apparently a number of inherent safety factors or mechanisms which decrease the amount of biologically available Sr^{90}. This speculation is further strengthened when one compares 1958 and 1961 Sr^{90} soil levels; there is no significant difference.

ATMOSPHERIC INFLUENCES ON FALLOUT DEPOSITION

PART 1. PARTICLE BEHAVIOR, SCAVENGING, AND DEPOSITION

Session Chairman: Joshua Z. Holland, Chief
Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission

RAIN SCAVENGING STUDIES*

A. Nelson Dingle
University of Michigan
Ann Arbor, Michigan

Introduction

The studies that we have been conducting under this title have as their broad objective the investigation and, as far as possible, the determination and quantification of the processes by which rain cleanses the atmosphere.

To accomplish this objective we have engaged in (a) a program of instrumentation designed to provide pertinent new types of observational data, (b) a collaborative program of controlled tracer scavenging experiments, (c) a program of rain sampling, and (d) theoretical developments designed to model the physical processes of scavenging in quantitative terms. In addition, during the year just past, we have moved our observing and sampling station to a new site at the Willow Run Laboratories, the University of Michigan. Except for (b) above, these activities are reviewed in the present paper.

The Willow Run Meteorological Field Station

Site description. The new site is located at the Willow Run Research Center of the University of Michigan, at the Willow Run Airport, about 12 mi. east of Ann Arbor. The Willow Run site is an open expanse of flat land providing a long wind fetch over grass. The breadth of the meadow available makes it possible to establish instrument sites that are therefore quite free from building-induced turbulence.

In the present setup three standard rain gases are placed, each at 100 yds. from the raindrop sorter in the north, east, and south directions, respectively. A tipping bucket rain gage is located 50 yds. east of the sorter, and is connected to an Esterline-Angus recorder located in the housing adjacent to the sorter. Fig. 1 is a map showing the location and distribution of instruments and facilities.

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* Publication No. 55 from The Meteorological Laboratories of the University of Michigan.

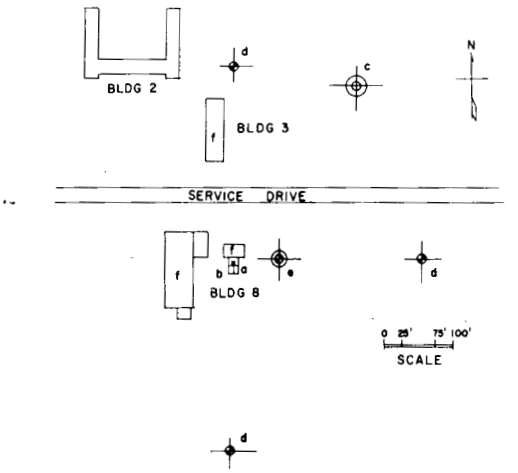


Figure 1. Schematic map showing relative locations of (a) the raindrop sorter, (b) the large sampling pans, (c) the raindrop-size spectrometer station, (d) the three standard rain gages, (e) the tipping bucket gage, and (f) the adjacent buildings.

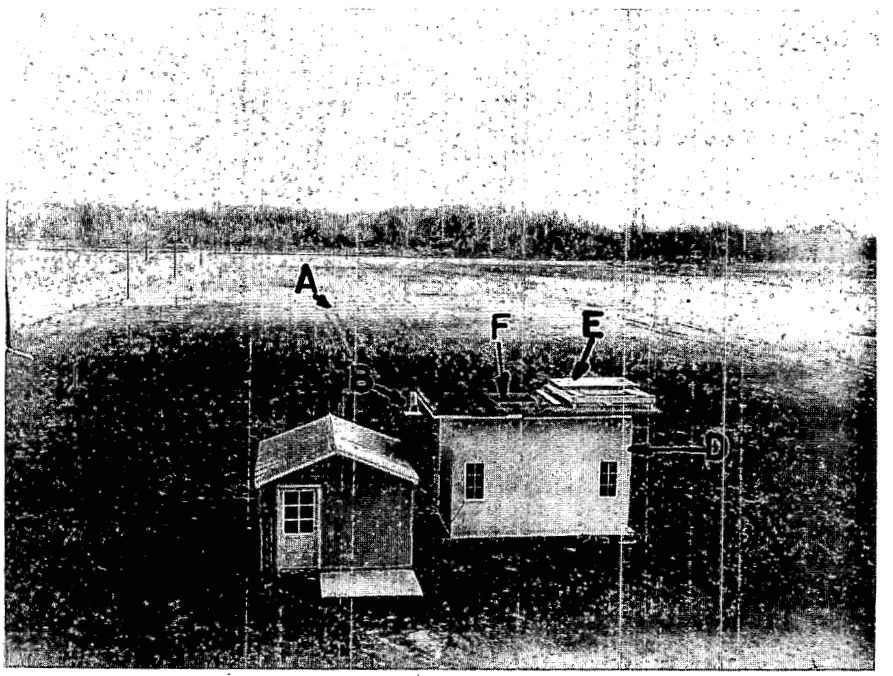
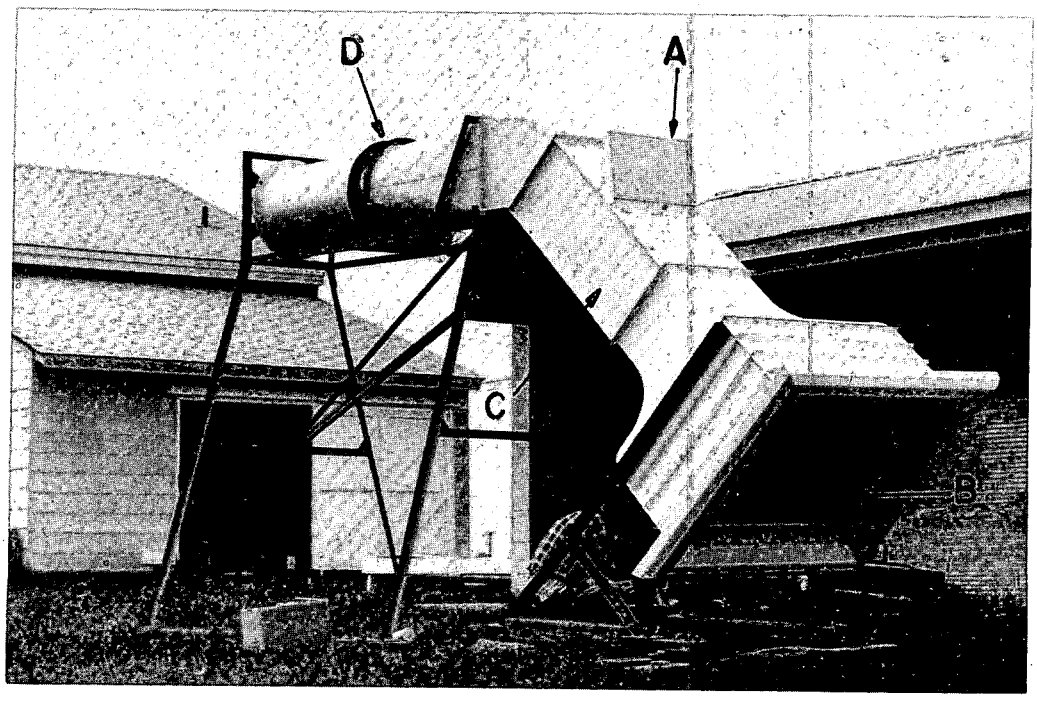


Figure 2. View of raindrop sampling station looking east, (a) standard rain gage, (b) tipping bucket rain gage, (c) recorder shelter, (d) raindrop sorter housing, note screening installed on roof to reduce rain splash, (e) large rain pans (one pan removed showing supports), (f) opening over sorter rain intake slit.



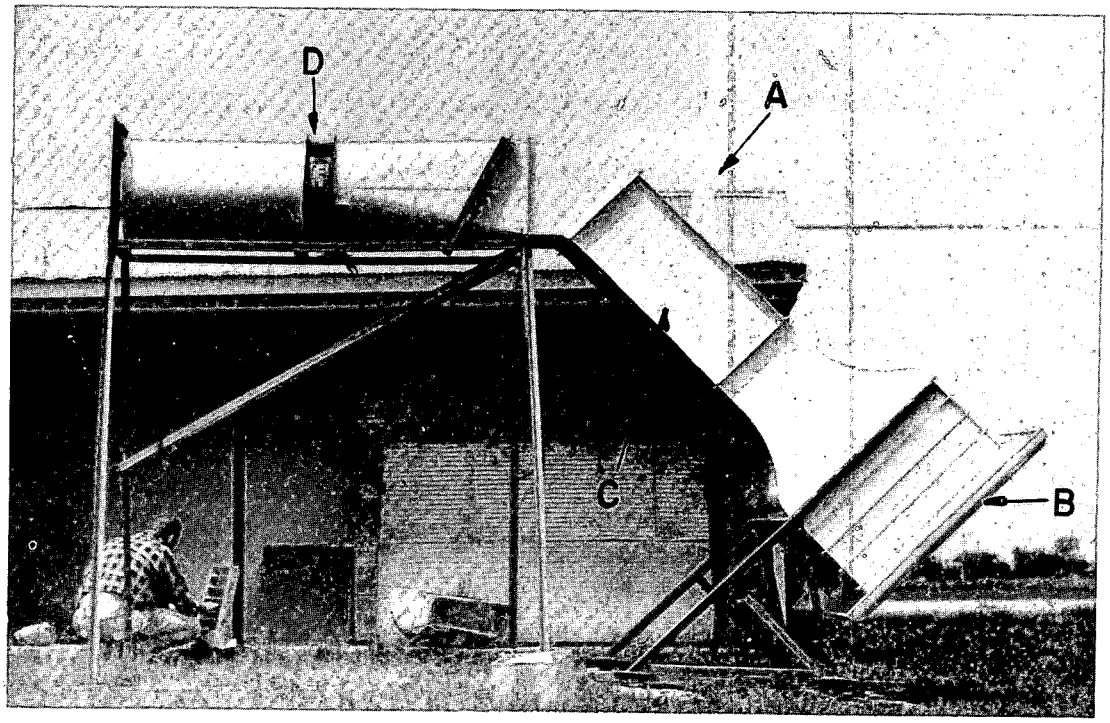


Figure 3. Photographs of the aerodynamic raindrop sorter: (a) rain sampling slit, (b) air intake, (c) drop sorting section, (d) fan assembly.

Because of the nature of the raindrop sorter, it was necessary to construct a housing around it to prevent the natural wind field from distorting the flow of air through the sorter. This housing is shown in Fig. 2. The large rain pans are placed on the roof of the raindrop sorter housing so that the rain flowing from these pans can be bottled and sealed by people working inside the sorter shelter for subsequent radiochemical analysis.

The raindrop-size spectrometer station is located roughly 150 yds. from the sorter housing in a northeasterly direction. The commitment of the raindrop-size spectrometer to our collaborative study with the Atmospheric Physics Research Group of the Hanford Atomic Products Operation has thus far made it impossible to station this instrument in its place at the Willow Run Field Site. As a result the data acquired during and since the summer of 1961 are not complemented by raindrop-size distributions.

Special instrumentation. The aerodynamic raindrop sorter has been described by Dingle (1). Photographs of the instrument as it was assembled at the Willow Run Field Site are shown in Fig. 3. The design is essentially a low speed wind tunnel the working section of which slopes upward at an angle of 45°. The latter criterion was determined by design studies which have been presented by Dingle and Brock (2). To prevent random air flow fluctuations through the working section, it is obviously necessary to isolate the air intake and exhaust from the free air stream. To do this the housing shown in Fig. 2 was constructed.

The large rain sampling pans, which together total 5.2 m^2 in area, have also beendescribed (2). These are located on the roof of the raindrop sorter housing so as to be as near its rain intake as possible, and to provide for reasonable convenience of the personnel attending the sampling units.

The tipping bucket rain gage has proved to be very useful in the documentation of amounts of rain and rainfall rates throughout rainstorms.

The raindrop-size spectrometer has, for the reasons explained above, not been in the field in compnay with the raindrop sorter and the rain collecting pans as yet. The need for this instrument in conjunction with the raindrop sorter, in order to make adequate analyses of the distribution of radionuclides in relation to drop size in rain is clear. Whereas the raindrop sorter must sample all through a given rain to acquire adequate samples, and thus it cannot distinguish samples according to time, the raindrop-size spectrometer gives the size distribution of drops throughout the storm (3, 4, 5) and thus provides data which allow for some interpretation of the time distribution of the water in the respective samples from the raindrop sorter.

An additional specialized piece of equipment which is capable of providing basic data for the study of the scavenging process is the zenith pointing radar. Steps have been taken toward renovating and modifying the Westinghouse MU-1 type marine radar to adapt it for use as a zenith pointing radar stationed at the rain sampling site. Accessory instrumentation required to record the

radar data include an oscilloscope for the presentation of the echoes and a scope camera suitable for the presentation of the echoes and a scope camera suitable for recoridng the scope images. The radar data thus obtained will provide vertical cross sections through the rain-producing systems as they move over the rain sampling station. Such cross sections will be related in a useful way to the sequence of drop sizes obtained by use of the raindrop-size spectrometer. Interpretation of the coordinated observations of raindrop-size spectra at the ground and of radar echoes of the storms in vertical cross section will be pursued on the basis of physical theory, and should lead to a much more adequate understanding of the rain process and hence also of the rain scavenging process.

Rain Sampling Results

During the month of September 1961, five of the rainstorms which occurred at the Willow Run Field Station yielded reasonably extensive sets of data.

The storm of 1 September 1961.

Examination of the synoptic situation shows that the rain which fell at Willow Run on this date was not associated with any frontal system whatever. The rain system originated as the result of an intensification of an easterly wave that progressed along the Gulf Coast and coverged with an isolated cold mass of superior air moving northeastward from the Texas area. This peculiar combination resulted in the development of a fairly extensive isolated precipitation area across northern Alabama and Mississippi, western Tennessee and eastern Arkansas and Missouri on the afternoon of 31 August. The surface trough associated with this rain system progressed northeastward and formed a weak low pressure center over Central Ohio by the morning of 1 September at which time the precipitation area extended over most of the States of Ohio and Michigan and northward into Ontario. There is, therefore, good evidence that no air mass change whatever took place on this occasion.

The rain began at 10:30 and continued until 13:13 EST. It was of showery character accompanied by occasional thunder and lightning and by highly variable winds. The three standard rain gages at the station showed respectively 1.99 in., 1.86 in., and 1.85 in. as the total mount of rain. The record of the tipping bucket rain gage was used to obtain the detailed information of Fig. 4. A total of 29 sequential samples were collected, and of these 9 samples were analyzed for their radioactivity. Graphs showing the results of the radiochemical analyses are presented in Fig. 5.

The procedure in collecting these samples, all from the large pans, was to fill successive 1-gal. polyethylene bottles, noting carefully the time of strating and ending each sample collection. The samples are related to the independent rainfall intensity measurements through the time record. The samples submitted for analysis in the present storm represent the time periods indicted by the marks above the rainfall curves of Fig. 4.

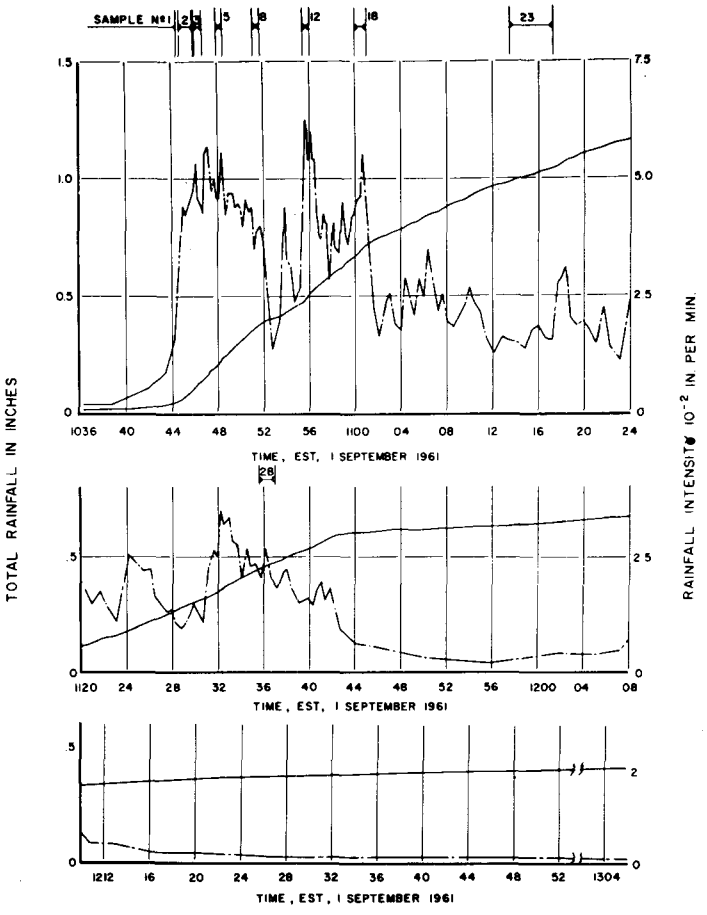


Figure 4. Rainfall amount and intensity from the tipping bucket rain gage record, 1 September 1961. Note that the timing of the samples submitted for analysis is shown above the precipitation curves.

The radiochemical analyses were performed by the National Sanitation Foundation located at the University of Michigan. An effort was made in this case to separate the filterable portions of the material brought down with the rain from the soluble portions and to analyze these separately. Although the technique is still in a stage of development and therefore is probably somewhat faulty, the findings appear to be quite interesting.

The filtering procedure is to use a series of graduated filters of the membrane type as supplied by Gelman*. The characteristics of the four filters that are used in this way are given in Table 1. The samples are filtered under pressure of about 100 lb./in.^{2}. As the first filter becomes clogged with material the pressure rises and the operator is warned that he must change the AM-1 filter. In some samples as many as three AM-1 filters have had to be used to complete the filtering process. As the data show (see Table 2 and Figs. 5, 7, 10, 12) the AM-1 filter catches by far the largest amounts of the filterable radioactive material. The AM-3 and AM-4 catch much smaller amounts and the AM-7 catches a somewhat larger amount than the -3 and -4 do. The principal reason for inserting the AM-3 and AM-4 filters in the sequence is to prevent the complete clogging of the very fine AM-7 filter. To eliminate short-lived nuclides, the samples are allowed to stand for 7 to 10 days before the analysis is performed.

* Gelman Instrument Company, Chelsea, Michigan.

TABLE 1

Characteristics of Gelman Membrane Filters

Type	Pore Size (μ)	Smallest particles retained with 99% efficiency (μ)
AM-1	5.0	1.0
AM-3	2.0	0.5
AM-4	0.8	0.1
AM-7	0.3	0.05

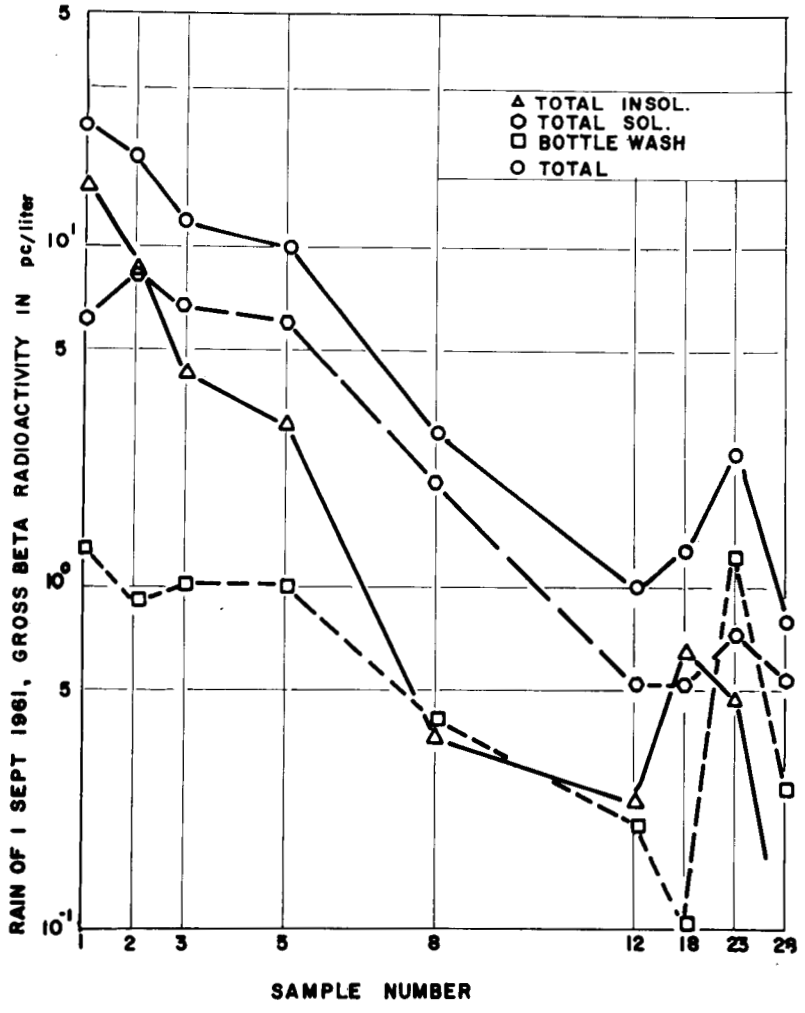


Figure 5. Gross β-radioactivity in rain samples of 1 September 1961. For the time distribution of the samples and their relation to rainfall intensity, see Fig. 4.

TABLE 2

Radioactivity of Sequential Rain Samples from Large Pan Collectors at Willow Run, Michigan

Gross Beta-Radioactivity pc/1

Sample Fraction

Identif. Date	#	Total Volume l	AM-1	AM-3	AM-4	AM-7	Total	Soluble	Bottle Wash	Total pc/l
19 VIII	1	4.10	9.8	1.2	0.8	2.2	14.0	8.3	-	22.3
	2	4.24	6.4	0.6	< 0.1	0.6	7.6	5.6	1.8	15.0
	3	2.08	5.7	0.6	0.6	0.4	7.3	5.6	0.6	13.5
24 VIII	1B	2.52	6.2	< 0.1	< 0.1	0.1	6.3	9.0	0.8	16.1
25 VIII	1	4.29	3.2	0.3	0.1	0.3	3.7	6.6	0.8	11.1
	2	3.88	2.8	0.5	0.3	0.4	3.9	7.6	1.0	12.5
1 IX	1	3.86	14.5	0.55	0.2	0.38	15.6	6.1	1.3	23.0
	2	4.02	6.8	0.97	0.23	1.4	9.3	8.3	0.9	18.5
	3	3.85	2.7	0.53	0.44	0.47	4.2	6.5	1.0	11.7
	5	4.03	2.2	0.39	0.16	0.22	3.0	5.9	1.0	9.9
	8	4.08	0.16	0.19	< 0.08	< 0.08	0.36	2.0	0.4	2.76
	12	3.79	0.23	< 0.09	< 0.09	< 0.09	0.23	0.5	0.2	0.93
	18	4.03	0.29	0.11	0.11	0.14	0.64	0.5	0.1	1.24
	23	4.04	0.16	< 0.08	< 0.08	0.3	0.46	0.7	1.2	2.36
	28	4.14	< 0.08	< 0.08	< 0.08	< 0.08	0.00	0.52	0.25	0.77
3 IX	1	4.06	5.0	0.39	0.08	1.0	6.4	12.0	0.9	19.3
	2	4.18	7.4	0.35	0.46	0.51	8.8	4.6	0.7	14.1
6 IX	1	4.12	4.3	0.35	0.35	0.3	5.3	6.2	0.53	12.03
	3	4.00	2.1	0.5	0.22	2.9	5.7	2.6	0.51	8.81
	5	7.83	2.0	0.20	0.21	0.59	3.0	2.2	0.65	5.84

(continued)

TABLE 2 (Continued)

Gross Beta-Radioactivity pc/l
Sample Fraction

Identif. Date	#	Total Volume l	Dissolved Solids mg/l	AM-1	AM-3	AM-4	AM-7	Total	Soluble	Bottle Wash	Total pc/l
12 IX											
	1	1.00	50.0	3.9	< 0.34	0.68	< 0.34	4.6	18.0	7.9	30.5
	2	7.71	18.6	4.0	0.16	0.16	0.13	4.5	7.0	1.7	13.2
	3	7.67	8.3	0.77	0.15	0.06	0.13	1.1	1.5	1.0	3.6
	5	7.88	10.8	0.71	0.09	0.10	< 0.01	0.9	1.4	0.4	2.7
14 IX											
	1	4.34	21.6	4.4	0.29	0.45	0.76	5.9	5.2	1.1	12.2
13 IX											
	1	3.65	25.4	147.	3.3	0.7	1.4	152.	130.	21.0	303.
	3	3.81	18.9	61.6	1.0	1.2	1.3	65.1	60.0	17.0	142.
	5	4.24	5.8	52.2	1.0	1.2	1.6	56.0	46.0	6.9	109.
	6	4.06	14.0	64.0	2.8	1.4	1.0	69.2	111.	10.0	190.
	8	7.84	5.5	18.0	0.6	0.5	1.4	20.5	31.0	7.5	59.0
30 IX											
	1	0.84	64.0	331.	10.0	6.0	55.0	402.	384.	97.0	883.
	2	0.90	67.0	316.	13.4	3.5	4.8	338.	310.	130.	778.
	3	3.69	24.4	286.	10.6	6.0	6.7	309.	330.	64.0	703.
	4	3.79	18.6	311.	5.3	2.6	4.2	323.	330.	76.0	729.
	5	3.96	17.2	290.	4.5	2.9	2.6	300.	420.	63.0	783.
	6	4.14	26.6	390.	2.4	4.4	3.3	400.	420.	65.0	885.
	7	3.86	31.0	350.	3.5	1.5	3.8	359.	400.	62.0	821.
	8	3.92	8.5	197.	5.0	0.9	2.3	205.	170.	13.0	388.
	9	3.62	12.5	31.0	3.2	5.5	10.5	50.2	64.0	38.0	152.

Returning to Fig. 5, note that the abscissa is the sample number on a linear scale out to No. 12. The data for the later samples are then plotted on a compressed scale to reduce the width of the figure. Whereas the sample number increases in time, it should be clear that it does not provide an accurate time scale. The timing of the respective samples in relation to real time (EST) and to the rainfall data is shown in Fig. 4.

The curves in Fig. 5 indicate that the insoluble particulates in the first sample of rain carried with them the majority of the radioactivity; however, by the time the second sample was collected the radioactive content of the insoluble portion was reduced to near that of the soluble portion, and in the third and succeeding samples, the soluble radioactive materials appear to have been quite predominant over the insoluble ones. The considerable departure of these data from a reasonably smooth decay curve suggests that fresh sources of contamination were drawn into the rain system during the course of its passage over the sampling station. It is clear, however, that by 10:56 when the 12th sample was taken, some 26 min. after the onset of the rain, the atmosphere in and under this rain system had been quite thoroughly cleansed of radioactive contamination. The period of rapid washout of accumulated debris thus appears to have been confined to the initial light rain and the first major shower (Fig. 4), but the 12th and later rain samples appear to have a uniformly low specific radioactivity (total gross β less than 2 pc/l). This includes the rain from the second major shower, that from the succeeding lesser showers, and also the steady gentle rain of the dissipating stage up to 11:55 EST when the last sample was closed off.

The fact that no air mass change, in the usual sense, occurred is especially significant. As the rain system progressed across the map, it evidently entrained fresh contaminated air into the leading rain-generating cells. Having processed this air through the leading shower mass, and thus having cleansed it, the system apparently reprocessed the clean air through the following shower masses adding contaimination to the later rain mainly by the entrainment of air from aloft, and finally it discharged clean air at its rear. A complex system of growing and decaying cells such as that proposed by Byers and Braham (6) for thunderstorms in suggested.

The storm of 6 September 1961. The synoptic character of this rain is not clear from the U. S. Weather Bureau daily maps, but the description of the rain as recorded in the project log book suggests the character of a squall line. The rain began at 15:27 EST and ended at 16:18 EST. Despite a dark and turbulent appearance of the sky there was very little lightning and thunder with this rain, and there were only relatively moderate winds. The total rain recorded by the rain gages was 0.37 in., 0.32 in., and 0.33 in. respectively. Again the detailed record of Fig. 6 was constructed from the tipping bucket rain gage record. A total of 6 samples were collected, and of these 3 were submitted for radiochemical analysis. The results of these

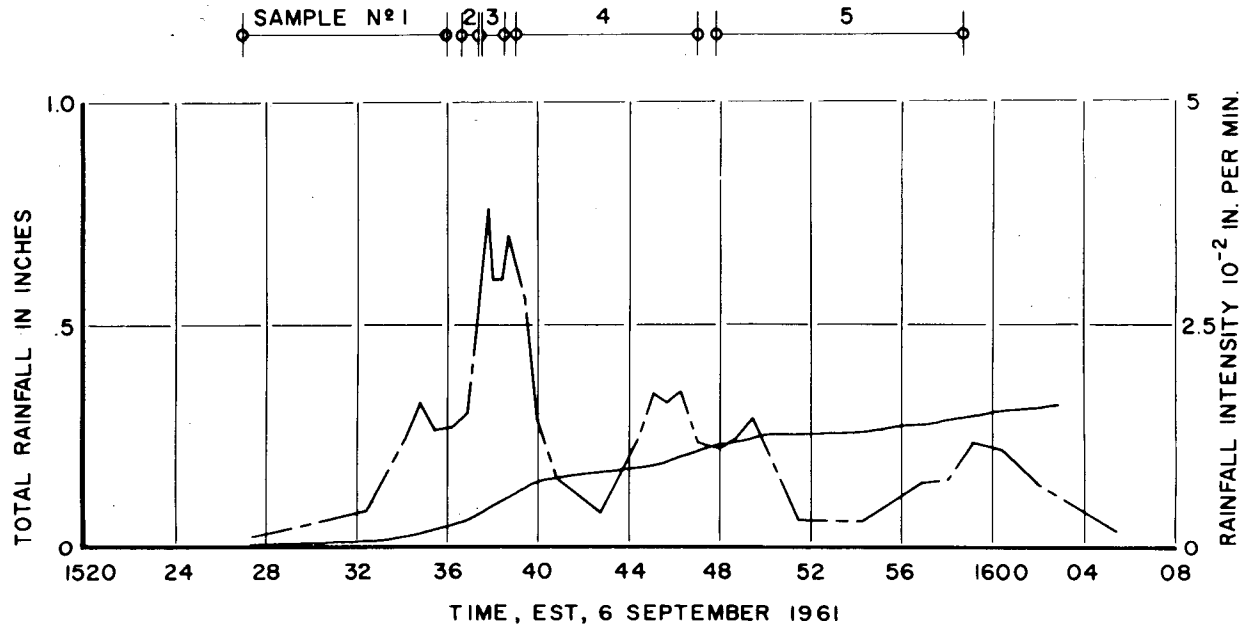


Figure 6. Rainfall amount and intensity from the tipping bucket rain gage record, 6 September 1961. The timing of the sequential samples is shown at the top of the figure.

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analyses are shown in the left part of Fig. 7. In this case the insoluble solids filtered from the rain carried with them the larger amount of radioactive material in the later samples, whereas the soluble material contained the larger amounts of radioactivity in the first sample. This is just the reverse of the relationship observed in the storm of 1 September. The fifth sample was taken about 1/2 hr. after the first one and in this time, the total radioactivity was reduced by a factor of about two.

The storm of 12 September 1961. The synoptic situation for this rain shows a stationary front in the vicinity of Willow Run with a strong flow of moist tropical air moving ahead of the remains of Hurricane Carla. The description of the rain on this occasion indicates that it was principally of the warm fron type, a light rain, more or less continuous, with some variation in intensity but not extreme variations. Winds were mainly from the east and southeast during the rain. The rain gages further bear out the uniformity of the rain in their readings which were 0.44 in., 0.42 in., and 0.43 in.

respectively. The tipping bucket record (Fig. 8) shows these characteristics in more detail. The rain lasted from 11:23 to 15:27 EST. Whereas the radioactivity associated with insoluble particles stayed about constant for the first two samples (Fig. 7), that of the soluble fraction started much higher and decreased very rapidly from the first to the second sample. After the second sample the specific radioactivity of the rain decreased in expected fashion to a minimum which prevailed for the third and fifth samples. The final level of specific radioactivity was of the order of 1/10th that of the first sample. [Ed. note: specific radioactivity" in this paper should read "concentration"].

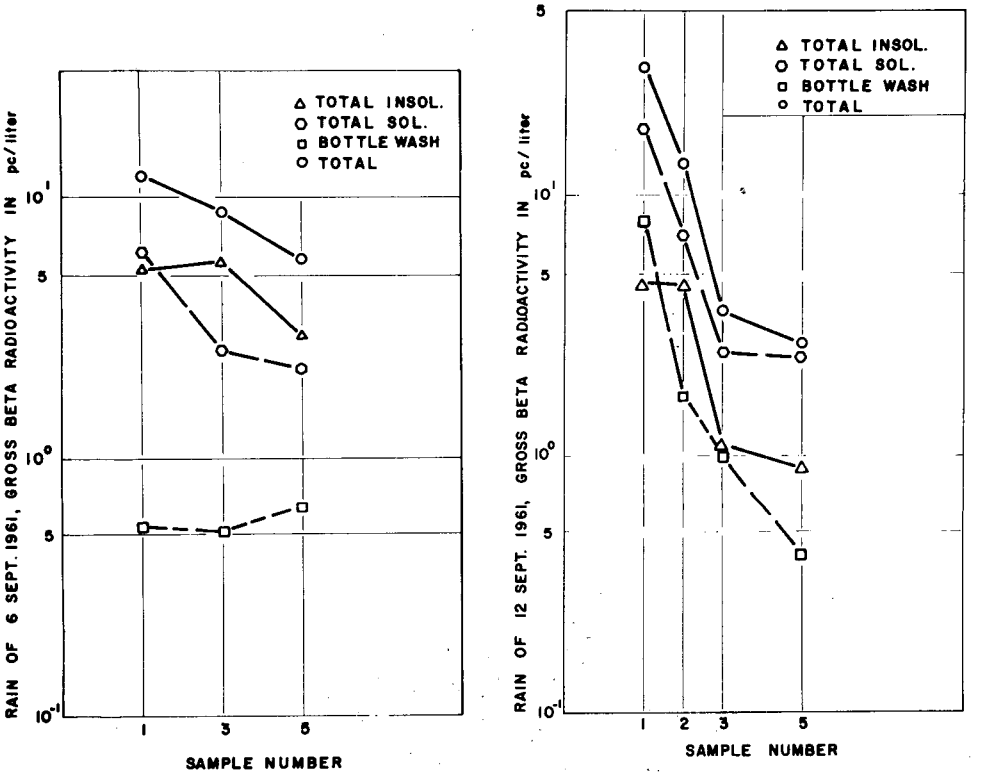

Figure 7. Gross β-radioactivity in rain samples of 6 September 1961 (left), and 12 September 1961 (right). For the time distribution of the samples and their relation to rainfall intensity, see Figs. 6 and 8, respectively.

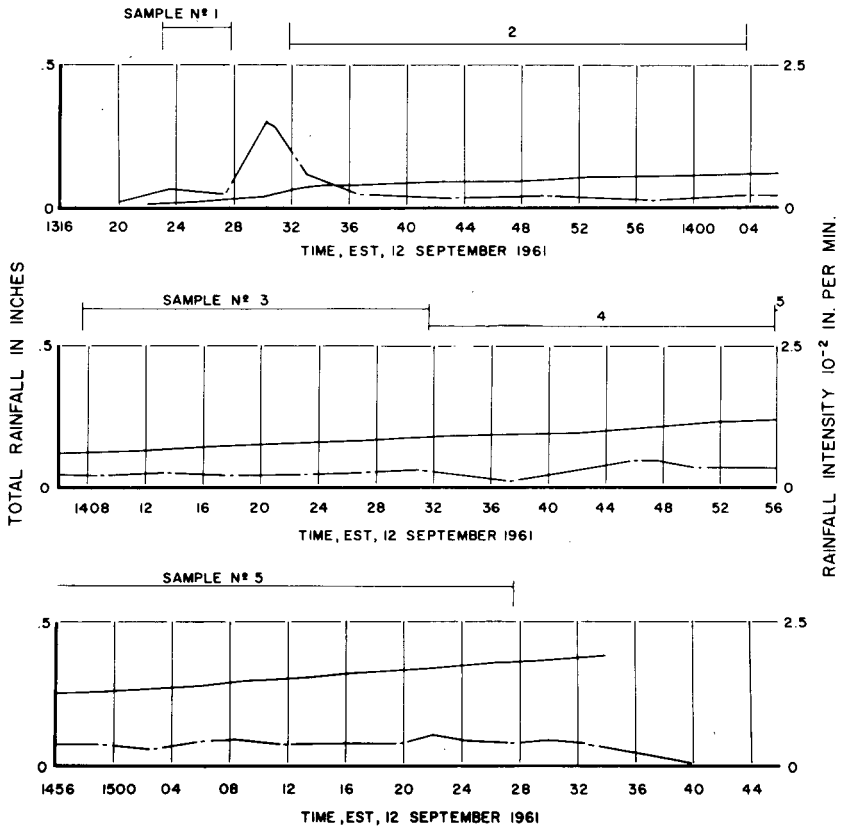

Figure 8. Rainfall amount and intensity from the tipping bucket rain gage record, 12 September 1961. The timing of the sequential samples is shown at the top of the figure.

The storm of 23 September 1961. The weather maps for the period of this storm show a cold front lying in a col which passed over sotheastern Michigan somewhat earlier than the recorded rain. The cold front was oriented roughly northeast to southwest and the ridge line through the col across southeastern Michigan was oriented at nearly right angles to the front. Rain sampling began at 20:39 and ended at 21:47 EST. Considerable lightning activity of the cloud-to-cloud type and general rumbling of thunder was observed during this storm. The total precipitation as recorded by the three rain gages was 0.76 in., 0.71 in., and 0.70 in. respectively. Fig. 9 shows the detailed results from the tipping bucket gage. Most of the rain fell in a short heavy thunderstorm which lasted from about 20.41 to 20:46 EST. The rain then stopped for a couple of minutes after which light rain was observed increasing slightly in intensity with time. A light and fairly steady rain persisted until the end of the storm. A total of 8 samples were collected, of which 5 were submitted for radiochemical analysis. These results are shown in Fig. 10.

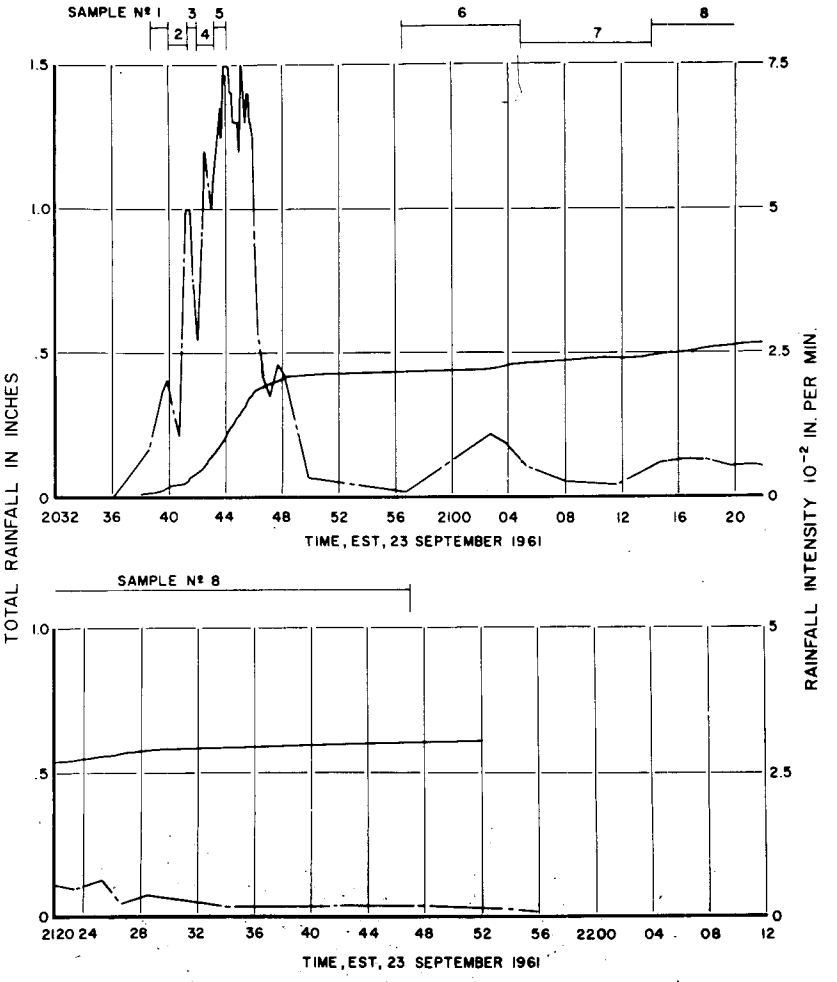

Figure 9. Rainfall amount and intensity from the tipping bucket rain gage record, 23 September 1961. The timing of the sequential samples is shown at the top of the figure.

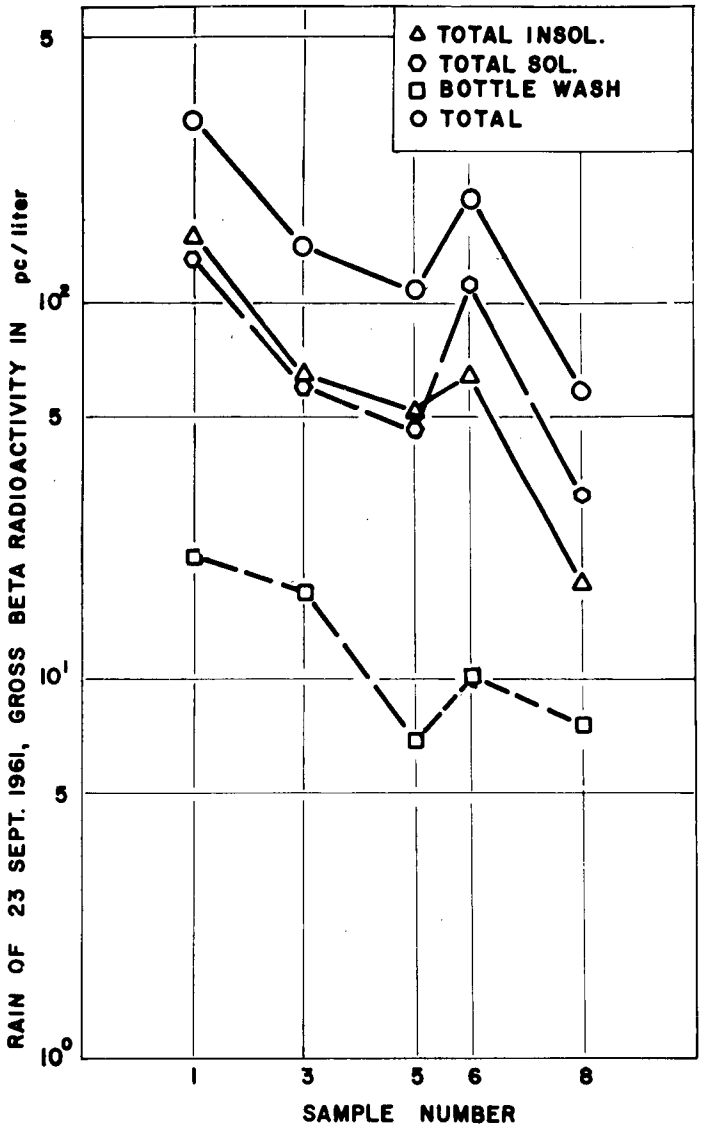

Figure 10. Gross β-radioactivity in rain samples of 23 September 1961. For the time distribution of the samples and their relation to rainfall intensity, see Figure 9.

A 10-fold increase of specific radioactivity following the resumption of atmospheric tests is clearly shown for the first time by these data. The first, third and fifth samples show the expected trend within the storm toward reduction of the specific radioactivity of the rain, but the sixth shows an abrupt increase. The eighth shows a sharp decrease. Interestingly, the period in which the major part of the rain fell (20:38 to 20:45) embraces the time during which samples 1 through 5 were taken. Samples 6 is therefore the first sample taken after the "couple of minutes" break in the rainstorm. It appears that an injection of contaminated air took place after the first heavy shower. This provides an interesting contrast with the storm of 1 September, and suggests that further study of these two situations might prove rewarding.

The storm of 30 September 1961. The storm system of this date was associated with a cyclonic storm which moved west and north of the area while a cold front extending southward swept from west to east across the area. The cold front was preceded by a squall line with which the most intense rainfall was associated. The rain began at about 21:00 and continued until about 23:00 EST. The rain amounts in this general area varied considerably in this case, one station in Ann Arbor reporting 1 in. of rain overnight while the Willow Run Station (12 mi. from Ann Arbor) accumulated only about 0.4 in. The amounts caught by the three rain gages were as

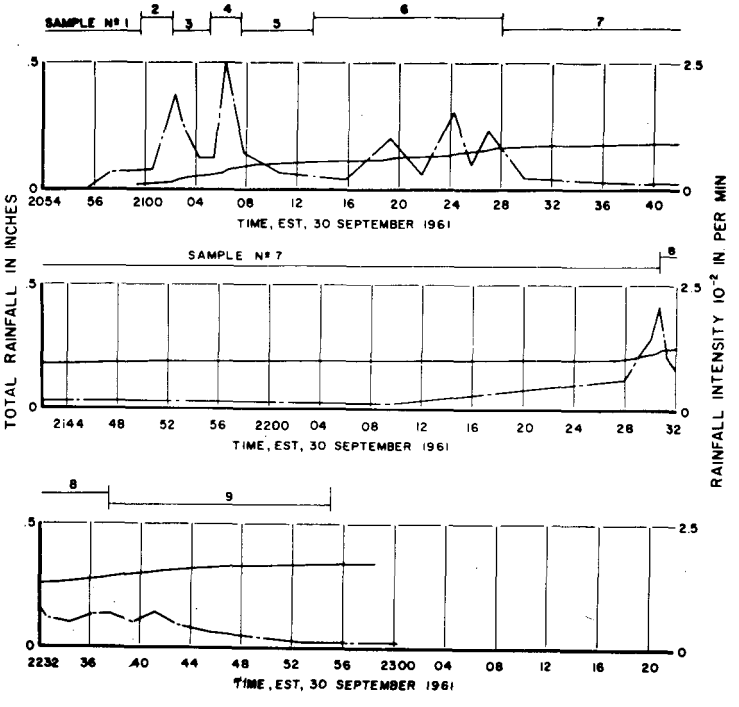

Figure 11. Rainfall amount and intensity from the tipping bucket rain gage record, 30 September 1961. The timing of the sequential samples is shown at the top of the figure.

follows: 0.46 in., 0.38 in., and 0.37 in. respectively. The storm was characterized by brief periods of no rain, of light rain, and of moderate rain. Details are shown in Fig. 11 derived from the tipping bucket record.

A total of 9 samples were collected and all of these were analyzed for radioactivity (Fig. 12). The changes of specific radioactivity again follow a peculiar pattern. The first three samples show a decreasing trend somewhat less pronounced than expected. The next three samples show an increasing trend to a maximum equal to the specific activity of the first sample. The last three samples then show a rapidly decreasing trend to the close of the storm. Throughout the storm the soluble and insoluble portions show about equal concentrations of radioactivity.

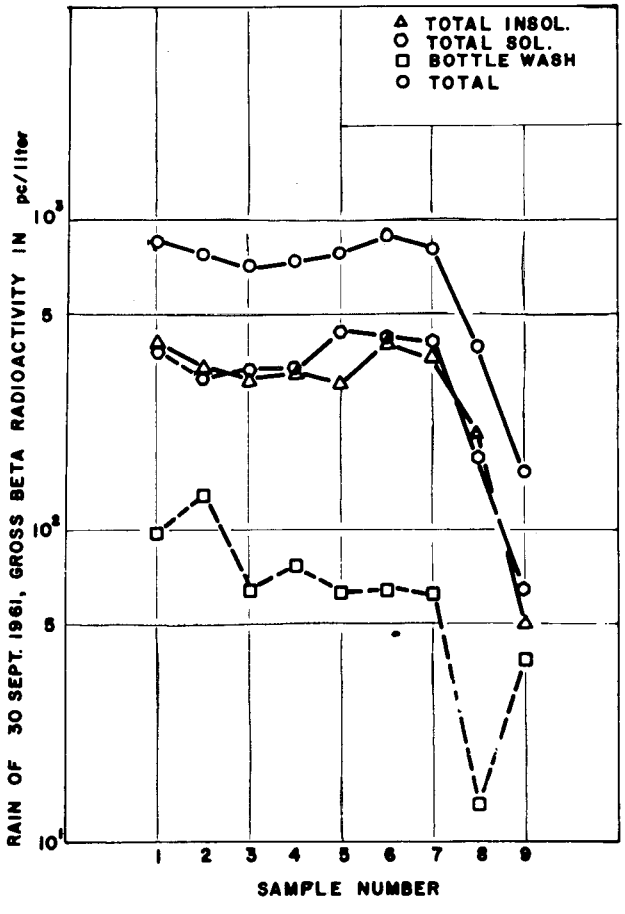

Figure 12. Gross β-radioactivity in rain samples of 30 September 1961. For the time distribution of the samples and their relation to rainfall intensity, see Fig. 11.

The trends of the specific activity curves for this storm suggests that the second band of showers, the rain from which is shared between samples 6 and 7, had access to fresh sources of nuclear test debris. In view of the limited rainfall intensities observed, it does not seem likely that the shower tops approached the tropopause. It is therefore probable that the contamination was entrained into the second band of showers with air from low levels. The nature of the rain, scattered showers as pointed out above, in itself suggests a spotty distribution of the air mass cleansing rather than a uniform cleaning of air over a broad area as in the storm of 1 September.

Further analysis of the 30 September 1961 samples. Because of the resumption of nuclear tests in the atmosphere and the consequent elevation

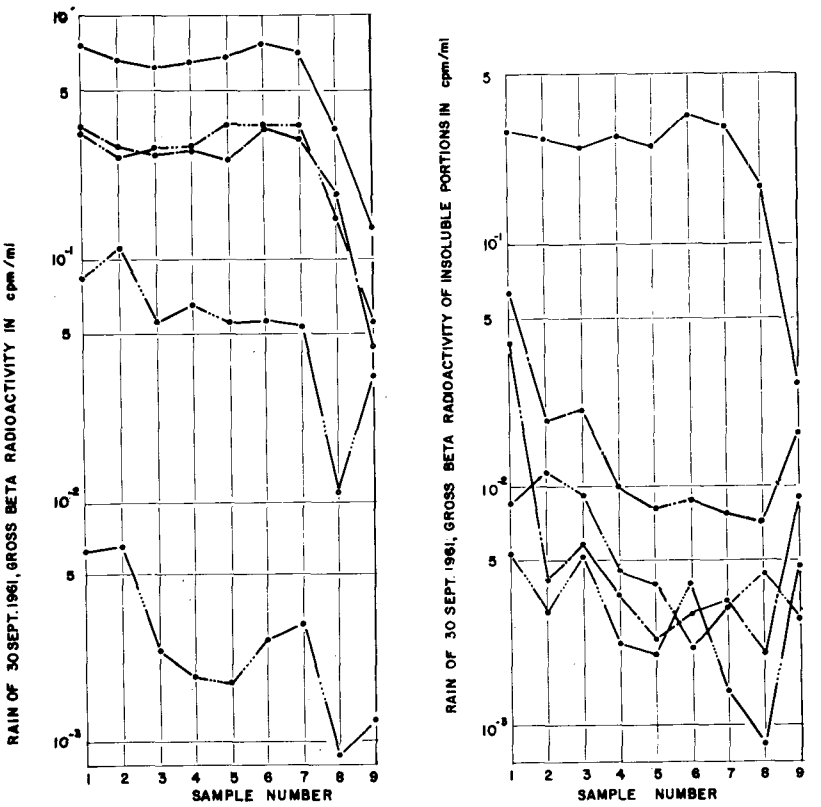

Figure 13. Details of the distribution of gross beta-radioactivity among the soluble and insoluble portions and among the respective filters. Left: lower curve, specific content of dissolved solids in mg/l; four upper curves duplicate the information of Fig. 12. Right: specific radioactivities of insoluble fractions caught on the AM-4 filter (lowest except for samples 6 and 9), the AM-7 filter (2nd lowest except for samples 1, 7 and 9), the AM-3 filter (3rd lowest except for samples 1, 6, 7 and 9), the sum of the AM-3, AM-4 and AM-7 filters, and the AM-1 filter (upper curve).

the radioactive contamination of the air, the specific radioactivity of samples taken on September 23rd and later was high enough to justify further study. In particular the samples taken on 30 September were studied in some detail as a pilot experiment. Fig. 13 shows on the left side an alternative from of the data contained in Fig. 12 and, in addition at the bottom of the figure, a curve showing the amount of dissolved solids in mg/l of rain. The interpretation of this curve is by no means clear but the upward trend of the dissolved solids observed in samples 6 and 7 shows an interesting paralledl with that of the total β radioactivity.

The right side of Fig. 13 shows a breakdown of the radioactivity found in the respective portions of the insoluble material. Clearly the AM-1 filter collects by far the large majority of the insoluble radioactive material. The next lower curve in this figure is the total of the radioactivity found on the 3 finer filter, the AM-7 shows higher radioactivity in the first and last samples than either the AM-3 or the AM-4. Although one might attempt to construct intricate interpretations of these observations at this point, it appears advisable to study in more detail the processes involved in the filtration and in the separation of the radionuclides.

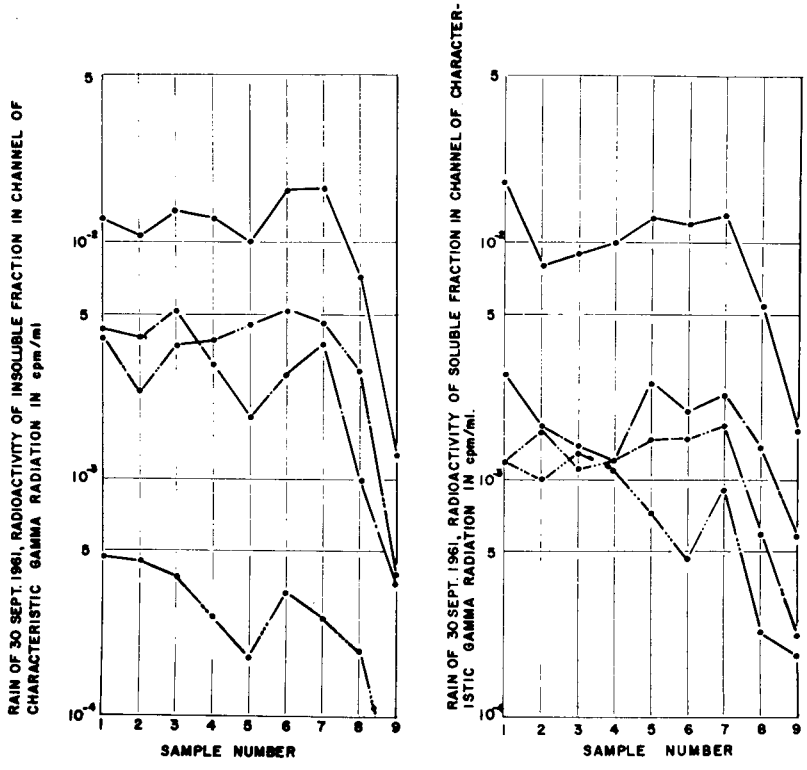

Figure 14. Results of gamma-spectrometric analyses. Left: The insoluble fraction. Upper curve, Ce^{141}, second curve (for samples 1, 2, 3), Ru^{103}; third curve (for samples 1, 2, 3), Zr^{95}; lowest curve, Ba^{140}-La^{140}. Right: The soluble fraction. Upper curve, Ru^{103}; second curve, Ce^{141}; third curve (except for sample 3), Ba^{140}-La^{140}; lowest curve (except for sample 3), Zr^{95}.

The results of gamma-spectrometric analysis of the soluble and insoluble portions, respectively, are shown in Fig. 14. The relationships of the specific activities among the respective nuclides of Ce^{141}, Zr^{95}, Ru^{103}, and Ba^{140}-La^{140} are very interesting within their respective fractions. The contrast between their concentrations in the soluble fraction as against the insoluble fraction are more so. Problems arise in the interpretation of these distributions because of the surface chemistry and physics and because of the ion exchange processes that probably accompany the filtration. It is therefore not clear that these curves truly show the relative proportions of the isotopes contained by soluble and insoluble materials. Further study of this kind of information would appear to be in order, and certainly more of this kind of data are needed before a firm interpretation can be presented.

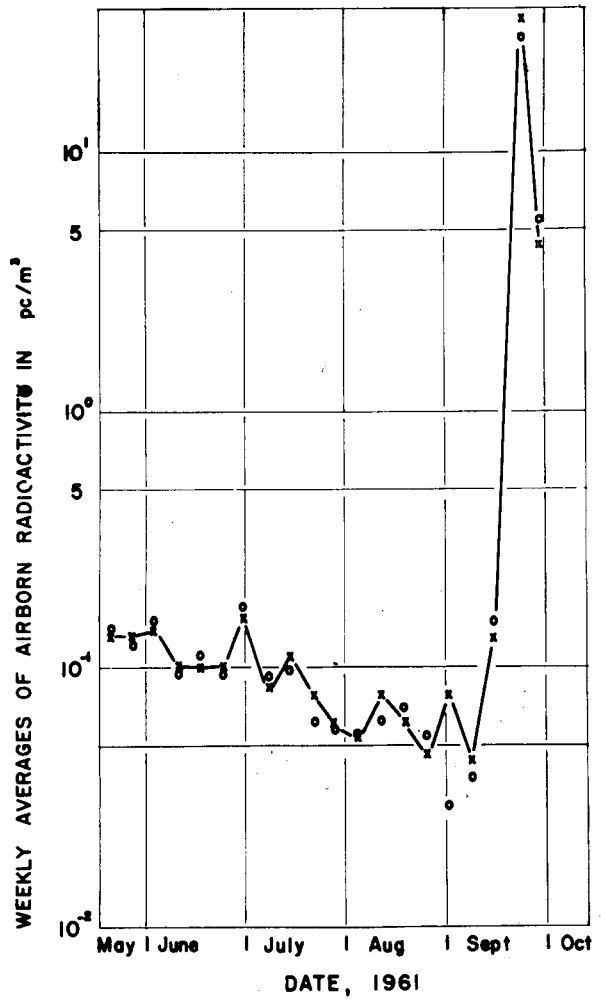

Figure 15. Weekly averages of gross β-radioactivity in airborne dusts collected at Ann Arbor and at Monroe, Michigan.

Air sampling data. The monitoring of airborne radioactive materials by the School of Public Health has provided additional data of value to our program. These data are taken from 2 stations in Ann Arbor and a third station at the Fermi Reactor Site at Monroe, Michigan. Fig. 15 shows the weekly averages of the airborne radioactivity from these 3 stations since May 1961. The observations taken on the Ann Arbor campus of The University of Michigan are joined by a line in this diagram. The principal purpose of this diagram is to show the dramatic increase of airborne radioactivity in mid-September.

Since the resumption of testing in the atmosphere the air monitoring program has been intensified by adopting a daily sampling period. The results of this program for the months of September and October are shown in Fig. 16. Here the upper curve gives the values of 24 hr. total beta counts of airborne dust and the lower curve gives 24 hr. values of I^{131} radioactivity in Ann Arbor. The daily samples are allowed to decay for 24 hr. before beta-analysis is performed in order to prevent distortion of the data by extremely short-lived nuclides and by natural radiation. This contrasts with the decay time of 7 to 10 days allowed for the rain samples.

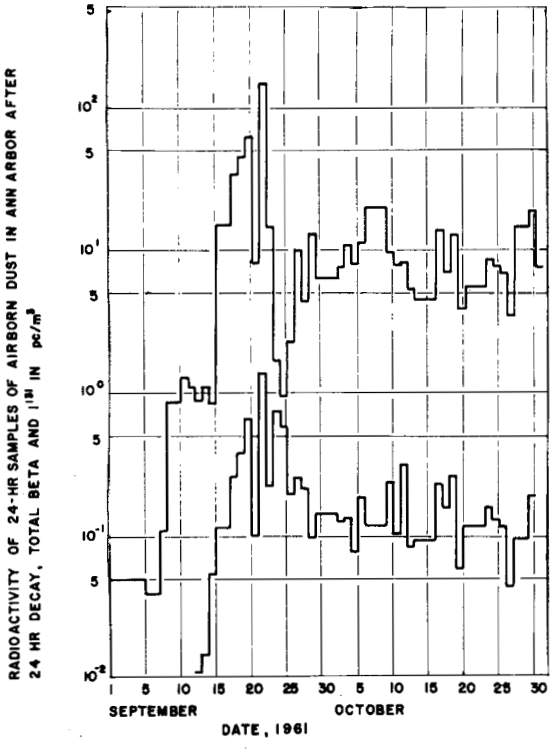

Figure 16. Total β-radioactivity and I^{131} activity in 24-hr. samples of airborne dust collected at Ann Arbor, Michigan.

It is of some interest to place these curves in juxtaposition to the data from the rain samples. The air samples show that the airborne gross beta-count increased somewhat more than 10-fold about 7 or 8 September and that it took another 10-fold jump on 15 September going an additional 10-fold higher still on 22 September, a total increase from the 7th to the 22nd of about 1000-fold. The airborne radioactive iodine, on the other hand, increased abruptly first on 14 September, and then by steps to a maximum 100 times the amount present on 10 September by 22 September. In contrast to these results we have already noted that the rain of 12 September showed no appreciable increase of radioactivity over the rains of the 1st and the 6th. In addition a light rain which occurred on 14 September also showed no increase of radioactivity. The first large increase observed in the rain-scavenged material was observed on 23 September and represented a 10-fold increase. This corresponds to the time when the airborne dust had increased 1000-fold in its radioactivity and the airborne iodine had increased about 100 fold. It is clear that some of the differences noted are attributable to short-lived radioactive nuclides. The fact that the rains of the 12th and 14th failed to show the 10-fold increase revealed by the atmospheric dust suggests that this increase in the dust was primarily attributable to the short-lived nuclides. What is not clear is the mechanism whereby the short-lived nuclides reached Ann Arbor in the air nearly two weeks before the large increase of longer-lived nuclides occurred.

Summary. Unfortunately the time available between the collection of these data and the submission of the present report has not been sufficient to make comprehensive analyses of the observations. The above superficial study of these data serves only to indicate some of the principal points of interest. There remain possible questions as to analytical technique, which is still being developed, and of course there are numberous facets of the data which deserve further exploration and study. Particularly in the realm of synoptic meteorology, more detailed investigation of the rains of 1 September and of 23 and 30 September are planned. It is anticipated that cooperation between our group and the National Sanitation Foundation group will lead to more complete studies of the radioactive debris contained in airborne dusts and of the relation of these data to the findings obtained from rain samples.

The Relationship Between Scavenging and Rain Generation

It is perfectly clear that the scavenging processes are intimately associated with the processes of rain-generation. It is therefore only logical that one consider the role of each of these processes throughout the development of clouds and the generation of rain in rain-producing storms. If one starts by considering what is a reasonable model of a rainstorm, it is possible that the scavenging processes might be brought into an improved perspective.

Any reasonable model of a rainstorm must start with the growth of cloud droplets on a spectrum of condensation nuclei which, in turn, yields the spectrum of cloud droplets. There is in existence much useful information on these specific points in the cloud physics literature. Following the initial phase of condensation, one must think of the coalescence of cloud droplets by collision processes and/or the rapid growth of cloud particles by an ice process, in order to accomplish the production of rain from cloud particles. Both the ice process and the collision processes have statistical characteristics which are subject to some reasonable expression, and again the reference to precipitation physics studies is appropriate. Having produced a spectrum of raindrops, one must think in terms of the variable fall speeds of the drops through lower portions of the cloud, through the cloud base, and thence to the ground with an evaporation process occurring between the cloud base and the ground.

In connection with each of these processes, the growth of the cloud droplets, the collection of cloud droplets into raindrops and the falling of raindrops through the air, the accompanying scavenging processes should be considered. These may be thought of as the Brownian diffusion of very small particles as suggested by Greenfield (7), the possible diffusion of intermediate size particles by microturbulence, and the collision and impaction collection suggested by the work of Langmuir (8).

But over all of these processes and controlling them throughout any storm, the principle of continuity must be considered. Because the system involves 3 basic categories of constituents, 3 interacting continuity equations must be invoked: 1) that for the air, 2) that for the water substance, and 3) that for the contaminant materials.

In studying the generation of radar echoes, Kessler (9, 10) has found the continuity equation to be extremely useful and to give results that agree very well with the experience of radar observers. At the University of Michigan we have undertaken to model the rain process in somewhat more detail, extending Kessler's continuity approach by accounting for the size spectra of cloud droplets, of raindrops, and of contaminant particles in integrating the rain-generating and scavenging processes layer by layer through a hypothetical atmosphere. This effort appears to hold a great deal of promise, and we plan to present further details and results in a future report.

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THE INTERACTION BETWEEN RADIOACTIVE AND NON-RADIOACTIVE PARTICLES IN THE STRATOSPHERE

James E. Manson
Air Force Cambridge Research Laboratories
Bedford, Massachusetts

Introduction

The purpose of this paper is to discuss the relationship between the particles which carry nuclear debris and the non-radioactive particles occurring naturally in the stratosphere. That a positive correlation exists will be demonstrated, and the importance of this relationship with respect to the history of nuclear debris in the stratosphere will be examined.

During the past year the Nuclear Studies Branch of the Air Force Cambridge Research Laboratories has prepared, among other contributions, a series of papers describing the vertical profiles and horizontal extension of non-radioactive "natural" aerosols in the stratosphere. The altitude range studied varied from 5 to 30 km. The radius of the particles studied ranged from 0.01 to 5 μ. The purposes of this study, together with the theory of behavior of aerosols in the stratosphere and a description of the various experimental techniques used in the balloon phase of these investigations are discussed by Junge, Chagnon and Manson (1). An extension of this work, including a more detailed consideration of the techniques of measurement of the concentration of the very small particles between 0.01 and 0.1 μ radius (Aitken Nuclei) was presented by Junge (2). A similar extension for the large particles between 0.1 and 1 μ radius was presented by Chagnon and Junge (3). The result of these balloon studies was to indicate that a deep layer of particles existed with a maximum in number concentration between 16 and 24 km. This layer was found to consist of sulfate particles with radii between 0.03 and 1.0 μ.

In order to determine the horizontal extent of this maximum in particle concentration, collectors were installed on two of the Lockheed U-2

aircraft operated for the High altitude Sampling Program of the Defense Atomic Support Agency. The results of analysis of samples so collected were presented by Junge and Manson (4). This work confirmed the fact that the predominant constituent of these particles was sulfate and proved that the concentration indicated at mid-north latitudes by the balloon work did indeed extend over the large fraction of the stratosphere sampled by the High Altitude Sampling Program U-2 trajectories at 18 to 20 km altitude. A summary of this work and a discussion of the possible origin of these particles in gas reactions in the stratosphere was presented by Manson, Junge and Chagnon (5).

Through the use of the information included in all these studies, together with studies carried out by Friend and Sherwood (6), one can develop a reasonably accurate model size and number distribution of non-radioactive sulfate particles at the 20 km level in the stratosphere.

Junge, Chagnon and Manson (1) have presented a series of size distributions measured at various altitudes in the stratosphere, and Junge and Manson (4) have discussed the connection between the size distributions and the derived mass distribution at this level in the stratosphere. As pointed out in the latter paper, the disagreement noted between the computed mass concentration and that directly measured by the chemical analysis could be resolved by assuming a uniform spreading factor of the particles as collected on the glass slides and electron microscope screens. It was shown that this colrection preserved the form of the distribution between 0.1 and 1 μ radius, that is, the distribution in this radius range remained inverse to the square of the radius when presented as a function of the log radius. An alternative correction to bring the measured and computed mass concentrations into agreement would be to change the form of the distribution, allowing the distribution to vary inversely as the cube of the radius. Independent of this work, Friend and Sherwood (6) had determined that the thickness of the particles as deposited on formvar-coated electron microscope screens remained constant independent of particle size and this thickness measured to be approximately 0.2 μ. Insertion of this correction into the inverse square particle distribution converts it immediately to an inverse cube distribution, and in this way the measured and computed mass concentrations can be brought into agreement. Further, the results of measurement of number concentration at 0.1 μ will be close to identical for these two distributions. Therefore for the present work we will choose as the model distribution for the ambient sulfate particles that shown in Fig. 1. It is clear from the above discussion that this model distribution is simply a reasonable, current estimate, taking into consideration all of the presently known measures of this population. For instance, the particles smaller than 0.1 μ between radius "a" and radius "b" in Fig. 1 have been added to the distribution in order to bring the total number concentration up to a value within reasonable agreement with the Aitken Nuclei measurements. An examination of the size distributions first reported by Junge, Chagnon and Manson (1) indicates that particles in this size range are present at 20 km.

The density of the aerosol calculated from this model distribution is approximately 1.0 x 10^{-14} g/cm3, assuming a condensed phase density of 2.0 g/cm3, whereas the measured aerosol density is 1.6 x 10^{-14} g/cm3. Considering the approximations involved in computing the collection efficiencies, determing the sample volume and estimating the composition, this is reasonably close agreement, and further refinement of the model is not necessary for our purposes at this time. Therefore, all of the computations in the present paper are based on the ambient number distribution shown in Fig. 1. The results of calculation will be compared to the measurements of total beta flux (count rate/counting efficiency) and sulfate density from a restricted group of impactor and filter samples collected between 30°N and 65°N between the dates of August 1960 and August 1961.

It should be indicated here that the results obtained by Friend and Sherwood (6) are in no respect limited to the measurement of deposited particle thickness, but encompass size distribution measurement, limited chemical analysis by electron diffraction and correlation of particle

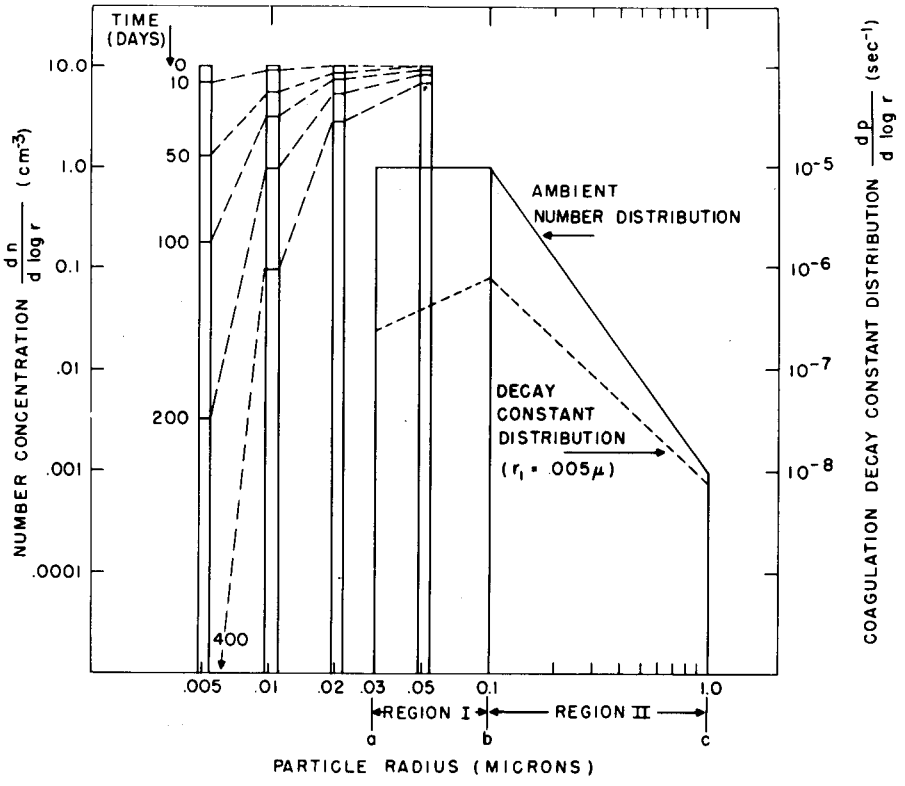

Figure 1. The coagulation of small particles into the ambient sulfare particle population. The progress of the coagulation is shown, together with the decay constant distribution as a function of radius for the smallest particles. Considering the sulfate population as a sink for the small particles, the decay constant distribution is actually a sink distribution.

volume density $\left(\int \frac{4}{3}\pi r^3 \frac{dn}{dr}\, dr\right)$ with filter Sr^{90} analysis. I have used only their thickness measurement in order to maintain as strict an independence of study as possible, and the remarkably close agreement between the aerosol density measured by these two approaches constitutes a confirmation of the validity of both.

Before proceeding to a discussion of the measurements it would be useful to review briefly the results of the theory of coagulation of small particles with the sulfate particles in the stratosphere.

II. Coagulation Theory

A brief treatment of the problem of coagulation of small particles into the ambient sulfate population has been presented by Junge, Chagnon and Manson (1). For the purposes of the present work, it is advisable to examine this question in slightly more detail. For derivation of eauations and discussion of the concepts involved reference is made to Zebel (7). The equation governing the number of small particles as a function of time (t) is given below, where for convenience of computation the size distribution is expressed as a function of radius (r) rather than of log r.

$$\frac{d\ln n_1}{dt} = -\frac{2kT}{3\eta}\int_a^c (r_1 + r_2)\left(\frac{1}{r_1} + \frac{\beta\lambda}{r_1^2} + \frac{1}{r_2} + \frac{\beta\lambda}{r_2^2}\right)\frac{dn_2}{dr_2}\, dr_2$$

$$= -\frac{2kT}{3}\left(\int_a^b \cdots dr_2 + \int_b^c \cdots dr_2\right) \quad (1)$$

$$= -(P_I + P_{II})$$

which yields: $$n_1(t) = n_1(0)\exp\left[-(P_I + P_{II})t\right]$$

where:

n_1 = number concentration of small (radioactive) particles;

n_2 = number concentration of ambient sulfate particles;

η = viscosity of air; $\beta \approx 1.5$ (Junge, Chagnon and Manson (1));

λ = mean free path; k = Boltzman's constant;

T = absolute temperature.

An examination of this equation indicates that the decay constant P is the natural function to consider in discussing the relationship between the rate of coagulation of small particles and certain features in the size distribution. In Table 1 we have presented the results of numberical calculations based on equation (1). Fig. 1 shows the ambient number distribution made up of region I and region II, and four vertical bars representing the various indicated sizes of radioactive particles which have initial concentrations arbitrarily chosen equal to 10. The concentration levels for these 4 sizes after a coagulation time of 10, 50, 100, 200 and 400 days is also indicated. The decay constant P is a measure of the rate of coagulation and thus the decay constant distribution given by dP/dr is a measure of the rate of attachment of the small particles with that part of the ambient number distribution lying betwenn r and r + dr. This function is plotted logarithmically in Fig. 1 for the smallest size fraction of radioactive particles and is seen to have a maximum at 0.1 μ for the assumed ambient number distribution.

TABLE 1

Coagulation decay constants and half-lives for various sized radioactive particles mixed with the ambient sulfate particle distribution

Radioactive particle radius r_1 (μ)	Decay Constants* Region I P_I (y^{-1})	Region II P_{II} (y^{-1})	Half-lives* $T_{\frac{1}{2}}$ (days)
0.005	9.0	5.9	17
0.01	2.3	2.0	60
0.02	0.73	0.40	220
0.05	0.29	0.082	690

*Calculated from equation (1) for 20 km altitude standard atmosphere and ambient distribution shown in Fig. 1.

III. Impactor and Filter Samples

The impactor samples used in the present study have been described previously by Junge and Manson (4), where they were referred to as tip samples. Since the form of the tip sample plate is not that of a simple ribbon, but is closer to a square prism, the impaction efficiency curves given by Ranz and Wong (8) cannot be applied simply. For this work we have chosen the average of the curves given by these authors for the impaction efficiency of a cylindrical impactor and a ribbon impactor. By doing this, we obtained the average impaction efficiency for the real aircraft direct flow rod impactor plotted in Fig. 2. This curve is calculated for a rod impactor 0.32 cm wide, exposed at 20 km altitude, flown at a speed of 770 km/hr., collecting particles of 2 g/cm^3 density. Also plotted on this figure are the ambient number distribution and ambient volume distribution curves, and the product of these curves with the impaction efficiency. The curves have been divided into region I and region II, and the integral of these curves is presented in Table 2. From this table we see that the impaction sample contains a small fraction of the number of particles in the ambient distribution, and a large fraction of the volume or mass of the ambient distribution. Fig. 2 shows also that this collector will not impact particles of this density smaller than 0.04 μ radius.

The filter samples studied in this work were collected on flights of the U-2 aircraft which collected the impaction samples mentioned above. These filter samples were collected on IPC 1478 paper described by Friend (9). Friend reported that, as applied in the U-2 sampling system, this paper was at least 95% efficient for the collection of radioactive particles in the stratosphere.

For total beta counting a samall disc was cut out of the large filter, from a position at about one half the radius from the center. Of this disc, a circular area of 15/16 in. diameter was exposed for counting, covered with mil mylar, following the procedure used by Isotopes, Inc. (9). The calculation of sample volumes thus follows a simple proportion to the total volume and total area for these filter samples.

The data pertinent to the collection of the impactor and filter samples used in this study are presented in Table 3.

TABLE 2

Comparison of ambient and collected number concentration and volume concentration from integration of curves in Fig. 2

Region	Number Concentration			Volume Concentration		
	Ambient $\Sigma \Delta n$ (cm^{-3})	Collected* $\Sigma \eta \Delta n$ (cm^{-3})	Ratio $\frac{\Sigma \eta \Delta n}{\Sigma \Delta n}$	Ambient $\Sigma \Delta \nu \times 10^{15}$	Collected* $\Sigma \eta \Delta \times 10^{15}$	Ratio $\frac{\Sigma \eta \Delta \nu}{\Sigma \Delta \nu}$
I	0.52	0.036	7%	0.6	0.07	11%
II	0.14	0.047	34%	4.2	2.7	.64%
I + II	0.66	0.08	12%	4.8	2.8	58%

* For the stratosphere at 20 km altitude and collection on a rod impactor 0.32 cm wide in an air stream velocity of 2.14×10^4 cm/sec (770 km/hr.), assuming spherical particles of density 2 g/cm^3. In this table and what follows we use η to indicate the impaction efficiency.

TABLE 3

Impactor and filter collection data

Sample Number.	Date Collected	Average Altitude (km)	Latitude Range (°N)	Longitude Range (°W)	Exposure Time (hrs.)	Path Length (km)	Ambient Volume (m3)	Reduced Volume (S.C.F.) *
1	25 Aug 60	20.0	32-38	100-121	5.02	3865	169	430
2	8 Sep 60	19.3	30-33	90-100	3.56	2741	120	340
3	4 Oct 60	19.7	30-44	99-111	5.5	4235	115	310
4	18 Oct 60	19.4	30-34	82-99	5.8	4466	201	570
5	27 Oct 60	19.8	32-64	101-144	6.0	4260	202	530
6	18 Jan 61	19.5	32	101	6.0	4620	213	590
7	13 Feb 61	13.7	41-45	93-101	3.0	2172	97	670
8	13 Feb 61	19.8	40-45	93-101	3.0	2310	102	270
9	27 Apr 61	19.9	33-64	102-146	6.0	4620	177	460
10	4 Aug 61	19.5	30	100	5.3	4081	206	570
5F (a)	27 Oct 60	19.3	37	103	1.57	-	-	118
5F[1](a)	27 Oct 60	20.2	60	104	1.35	-	-	87
6F (a)	18 Jan 61	19.9	32	111	0.95	-	-	55
8F (b)	12 Feb 61	20.5	38	105	1.6	-	-	90
9F (b)	28 Apr 61	19.1	38	104	1.8	-	-	137
10F (a)	4 Aug 61	19.5	30	-	-	-	-	93

(a) These filter samples were collected on the same missions as the impactor samples of the same numerals.
(b) These filter samples were collected on different missions from the impactor samples of the same numeral.

* S.C.F. standard cubic feet

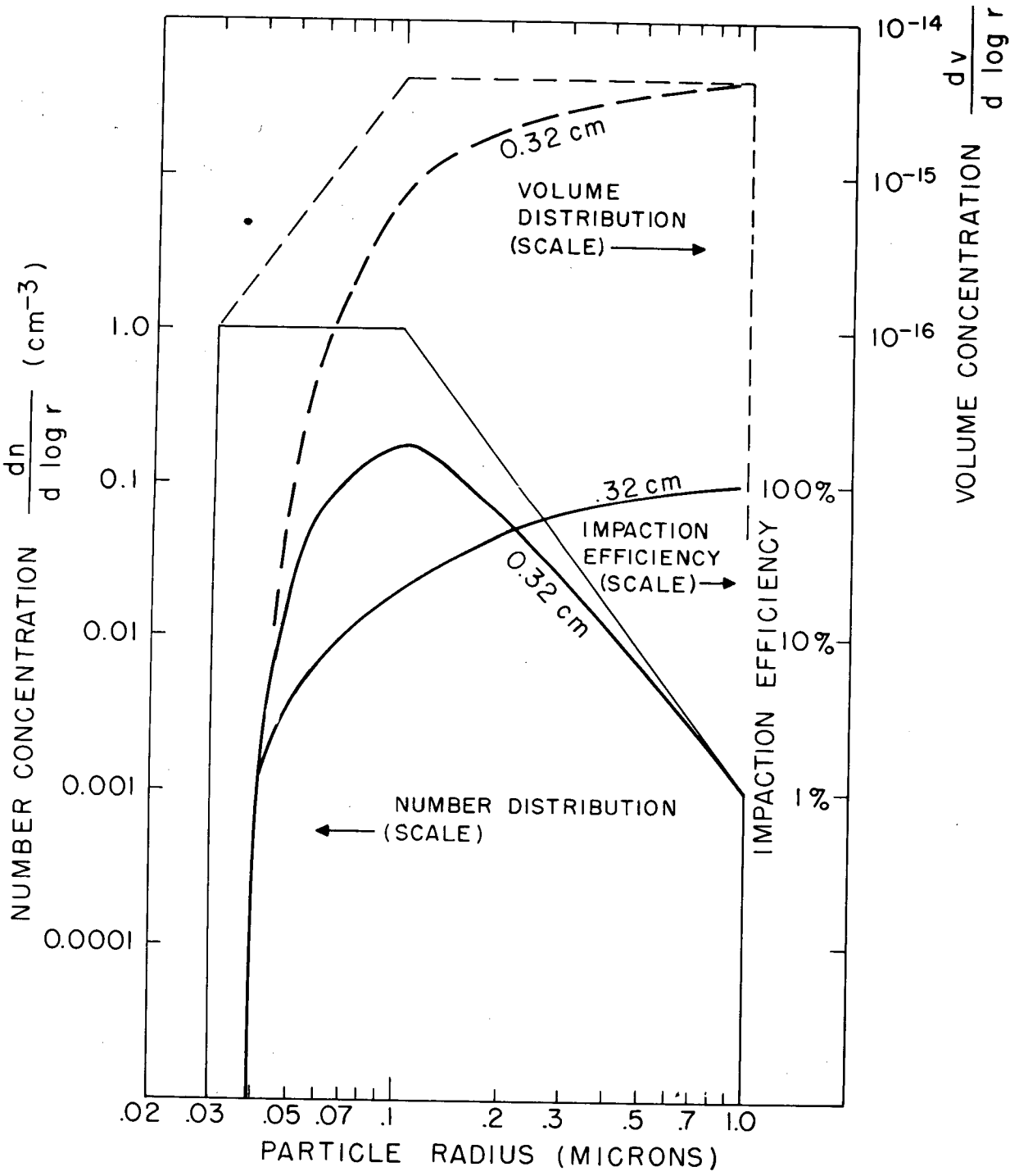

Figure 2. The ambient sulfate particle number and volume distribution, the impaction efficiency (η) as a function of radius and the product of the ambient distributions and the impaction efficiency to yield the collected number and volume distributions. The impaction efficiency and product curves are identified by the impactor diameter, 0.32 cm, and refer to collection of spherical particles of 2 g/cm^3 density in a stream velocity of 2.14 x 10^4 cm/sec at 20 km altitude.

An arbitrary numbering system has been used to identify the impactor samples as an aid in interpreting the data presented later as scatter diagrams. The numbering of filter samples follows that of the impactor sample collected on the same or nearest date. A significant difference between the exposure of the tip impactor samples and the filter samples is that the impactor samples are exposed for as long as 6 1/2 hrs. whereas the filter sample maximum exposure in this series was 108 min.

IV. Analytical Procedures

The techniques of non-radioactive chemical analysis of the aircraft impactor samples have been described previously by Junge and Manson (4). As an indication of the reliability of the x-ray flourescence technique of microanalysis for sulfur in these samples we present Table 4, which contains the comparative measurements on a pair of samples collected on 13 February 1961. These samples were collected on pieces of pure aluminum. The background readings refer to the counts obtained by analyzing a sample of the substrate aluminum of the same size and composition, but which had not been flown.

TABLE 4

X-ray flourescence analysis of two impactor samples for sulfur

Sample	Counts	Time (sec)	I (gross) (cps)	I (net) (cps)	m_s (mg)
Blank	200	1886	0.106	-	0
Standard	800	372	2.15	2.04	1.5
No. 7	400	2734	0.146	0.04	0.03
Standard	400	184	2.17	2.06	1.5
No. 8	400	1012	0.395	0.29	0.20
Blank	200	1956	0.102	-	0
Standard	400	175	2.29	2.22	1.5
Blank	200	2839	0.07	-	0
Standard	400	188	2.13	2.06	1.5
No. 8	400	1071	0.373	0.30	0.22
Standard	400	189	2.12	2.05	1.5
No. 7	200	1487	0.134	0.06	0.05
Standard	400	179	2.23	2.16	1.5
Standard	400	551	0.726	0.65	0.5
Blank	200	2547	0.079	-	0
No. 7	400	3192	0.125	0.05	0.04
Blank	88	1680	0.052	-	0
No. 8	400	1043	0.384	0.305	0.24

The standards were prepared by adding sulfur in the form of ammonium sulfate to blank aluminum slides insuring that the sulfate was restricted to the area being analyzed by the x-ray beam.

These data were taken with a vacuum spectrometer using an EDDT crystal, tungsten primary radiation at 50 kvp and a flow proportional counter detector with a nuclear type linear smplifier and pulse height analyzer set with the window width 25% of the base line. The quantity m_s is the mass of sulfur on a central area of the specimen 1.0 cm x 0.32 cm.

The results of analysis of all of the impactor samples are given in Table 5. It is noted that the values given for the density of sulfare in the stratosphere, ρ SO_4, has been corrected for the 58% volume collection efficiency of the impactor. For this reason the values given here are higher than those reported by Junge and Manson (4).

The procedures for obtaining the total beta activity measurements were chosen to insure simplicity and reproducibility. A Nuclear-Chicago automatic sample changer and scaler was used with a Nucleonics Corporation of America IWAA end window counter. This counter had a background count rate of approximately 20 cpm. Comparison standards were prepared using a National Bureau of Standards calibrated Tl^{204} standard. All of the tip impactor samples were counted sequentially with standards and blanks interleaved and at least 10,000 counts per sample were accumulated. In cycles of 2,000 counts per sample, the variation in rate from cycle to cycle was generally within $\pm \sqrt{n}/t$, where n is the counts per sample and t the counting time. The compilation of this counting data is given in Table 6 for all the impactor and filter samples used in this work.

The first two samples noted as Tl/Al and Tl/Ge are standards prepared on the aluminum and germanium substrate. As noted in the last column the values for these two standards are given in dpm and refer to the disintegration rate corrected for decay to 20 October 1961. It is seen in the fifth column of this table that the standard prepared on the germanium substrate has a higher net count rate than the other standard. Since one of our samples (No. 6) was collected on a germanium substrate it was necessary to include this standard in our counting sequence. The values listed under total beta flux for samples 1 to 10F are the product of the net counting rate and the dpm to cpm ratio. This ratio was formed from the data for the Tl/Al standard for all the samples except No. 6. For No. 6 the dpm to cpm ratio for the Tl/Ge standard was used to form the total beta flux product. Our counter was intercalibrated with the system used by Isotopes, Inc. (9) by exchanging impactor sample No. 9 and filter sample 10F. Isotopes, Inc. obtained net count rates for these samples of 14 and 24 cpm respectively, on 2 November and 26 October 1961.

TABLE 5

Results of sulfur analysis of impactor samples

Sample	σ_S ($g/cm^2 \times 10^{-6}$)	m'_{SO_4} ($g \times 10^{-6}$)	ρ_{SO_4} ($g/cm^3 \times 10^{-14}$)	ρ_{SO_4} ($\frac{g \times 10^{-9}}{S.C.F.}$)	ρ_{SO_4} ($\frac{g \times 10^{-7}}{Std.\ M^3}$)
1	1.11	1.83	1.5	5.9	2.1
2	0.62	1.02	1.2	4.2	1.5
3	0.84	1.02	1.0	3.9	1.3
4	0.50	0.85	0.57	2.0	0.71
5	1.07	1.76	1.2	4.5	1.6
6	1.70	3.00	1.9	6.9	2.4
7	0.14	0.23	0.35	0.51	0.18
8	0.80	1.32	1.8	6.8	2.4
9	1.36	1.95	1.5	5.8	2.0
10	0.55	1.05	0.73	2.6	0.93

Note: σ_S is the surface density of sulfur on the center of the impactor sample plate.
m'_{SO4} is the total mass of sulfate ion in the entire impactor deposit.
ρ_{SO4} is the derived density of sulfate in the air, given in three common units, calculated from σ_S, the path length and the volume collection efficiency.

V. Results

In Fig. 3 we have plotted the total beta flux per sample against the mass of SO_4 per sample. A positive correlation is evident. As demonstrated by Junge and Manson (4) the mass of SO_4 is a good measure of the total mass of non-radioactive material in the stratospheric aerosol, and it is as such a measure that we use it here.

The two pairs of points 3-4 and 7-8 require special note. Sample 3 was flown with two 1/8 in. diameter electron microscope grids attached in the center of the collection area, thus reducing the geometrical area and therefore the volume of this sample, since these grids were removed prior to the beta counting. From Table 5 we see that the density of sulfate in the stratosphere during collection of sample 3 was almost twice that during collection of sample 4, and that this more than compensated for the reduction in area of sample 3. In other words, sample 3 was collected from a smaller volume of air, but had a higher mass of sulfate than sample 4. It is significant that the total beta activity followed the sulfate in this, so that a positive correlation exists between these two variables for this pair of samples.

As indicated in Table 3, sample 7 was collected at low altitude and sample 8 was collected at high altitude. It is interesting that these two points follow the average correlation, since the decrease of both variables with altitude probably is due to diffusion and washout to the troposphere. The indicated correlation is exactly what one would expect if the particles carrying the sulfate and those carrying the activity are identical at the sampled altitudes.

One might suggest that the correlation indicated for the total quantity variables in Fig. 3 could be caused by the common factor of sample volume. This was discussed above in considering the pair 3-4, which indicated the opposite effect. Further proof that this is not the case is contained in Fig. 4, which presents the total beta flux per sample plotted against the reduced sample volume. A negative correlation is seen to exist between the two pairs of special samples 3-4 and 7-9 on this plot, and there seems to be no significant correlation for the group as a whole. The same is indicated by a plot of total beta flux versus ambient sample volume.

The possibility of a seasonal trend causing a spurious correlation is eliminated by a consideration of Fig. 5, in which the activity density is plotted against the sampling date. It must be remembered that these total beta counts were made on 20 October 1961 (Table 6).

Up until now we have considered only the correlation of various measures of the impactor samples. As indicated in Table 3, we also had available six filter samples from five flights during the same sampling period. Samples 5F and 5F' were collected at the beginning and end of the same trajectory over which impactor sample 5 was collected. Sample 6F was

TABLE 6

End window beta counting data for impactor and filter sample

Sample	Total counts	Time (min)	Gross rate (cpm)	Net rate (cpm)	Total β flux cpm (or dpm*)
Tl/Al	16,000	47.4	337	317	979*
Tl/Ge	16,000	40.4	396	376	979*
1	16,000	339	47.1	26.8	82.8
2	16,000	432	37.1	16.8	51.9
3	16,000	377	42.5	22.2	68.6
4	16,000	455	35.2	14.9	46.0
5	16,000	355	45.1	24.8	76.6
6 (Ge)	16,000	261	61.2	40.9	106.7
7	14,000	559	25.0	4.7	14.5
8	14,000	371	37.8	17.5	54.1
9	14,000	369	37.9	17.6	54.4
10	14,000	484	28.9	8.6	26.6
5F	16,000	213	75	55	164
5F'	16,000	247	65	45	134
6F	16,000	343	47	27	80
8F	16,000	245	65	46	135
9F	16,000	218	74	54	159
10F	14,000	246	57	37	110

* These values, given as dpm are calculated from the known properties of the standard solution.

**Impactor samples counted 20 October 1961; filter samples counted 10 November 1961.

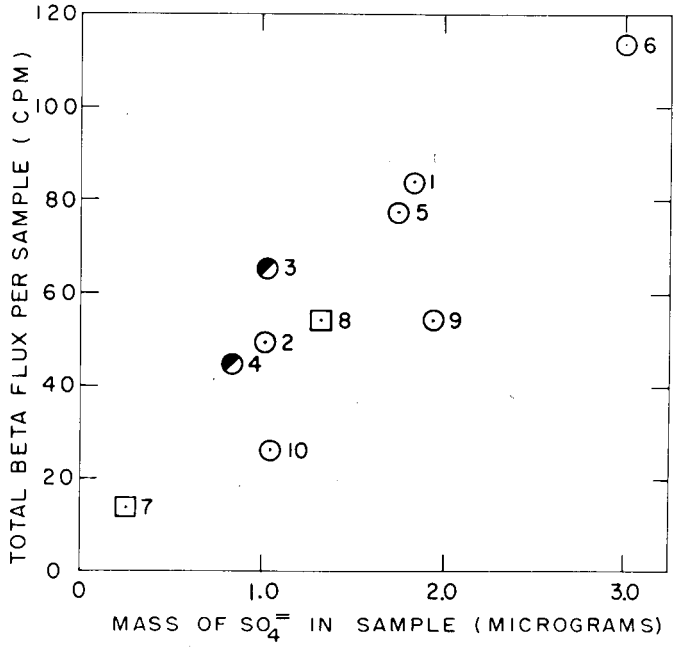

Figure 3. Scatter diagram of total beta flux (count rate/counting efficiency) per sample versus total mass of sulfate per sample. A positive correlation is indicated.

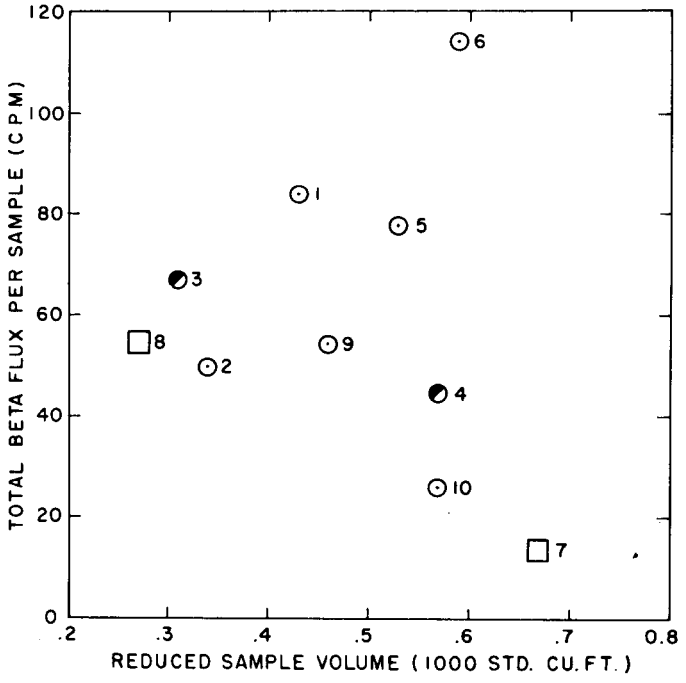

Figure 4. Scatter diagram of total beta flux per sample versus sampled volume at standard temperature and pressure. No correlation is indicated.

collected on the same trajectory on which sample 6 was collected. Sample 10F was collected on the same flight as sample 10. Samples 8F and 9F, as indicated by the collection date, were collected on different flights from impactor samples 8 and 9. However, other than a difference in date of one day, these samples were collected over very similar trajectories as the corresponding impactor sample. The filter samples are known to collect at least 95% of the trotal radioactive fraction of particles in the stratosphere at 20 km altitude. Therefore, a comparison of the collected total activity density on the impactor samples with the total activity density on the corresponding filter samples will indicate the fraction of the total activity collected by the impactor samples. This fraction is indicated in the last column of Table 7 and is seen to average 10% of the activity. The fourth column of Table 7 contains the ratio of the total activity per unit volume to the mass of sulfate per unit volume.

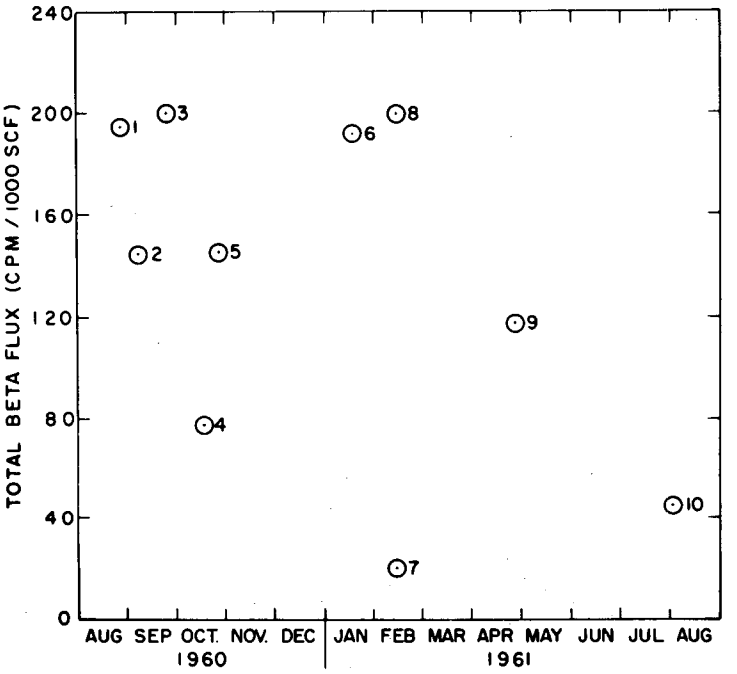

Figure 5. Scatter diagram of total beta flux density versus collection date. If there is any correlation here, it is very weak compared to that shown by Fig. 3.

TABLE 7

Comparison of filter and impactor samples

Sample pair	(A) Filter total beta density (cpm/SCF)	(B) Sulfate density ρSO_4 (g/SCF x 10^{-9})	A/B ($\frac{cpm \times 10^9}{g}$)	(C) Tip impactor total beta density (cpm/SCF)	C/A
5 - (5F, 5F')	1.5	4.5	0.33	0.15	0.10
6 - 6F	1.4	6.9	0.20	0.19	0.14
8 - 8F	1.5	6.8	0.22	0.20	0.13
9 - 9F	1.2	5.8	0.21	0.12	0.10
10 - 10F	1.2	2.6	0.46	0.045	0.04
Average	-	-	0.28	-	0.10

Thus, the total beta activity of an impactor sample has been found to be directly related to the total mass of non-radioactive material collected on the sample. The impactor sample, which contains 12% of the number of non-radioactive particles and 58% of the volume of non-radioactive material, has been found to contain an average of 10% of the total beta activity present in the sampled air. Further, as analyzed in October-November 1961, a specific total beta flux per unit mass of non-radioactive material in the stratosphere can be calculated, and is found to be 2.8×10^8 cpm/g. With an estimated conversion factor of 5% for Sr^{90} dpm/total beta cpm, this is in agreement with the value of 1.5×10^7 dpm/g found by Friend and Sherwood (6) for the ratio of Sr^{90} concentration to aerosol mass concentration.

VI. Conclusions

It is evident that with such a restricted number of samples, we cannot hope to prove the correlations indicated in the previous section. We can, however, examine the possible mechanisms of interaction between radioactive and non-radioactive particles at 20 km altitude in the light of these correlations.

It seems reasonable to assume from the data available at this time that the sulfate aerosol is a world-wide phenomena and that it is formed at approximately 20 km by photochemical oxidation of gaseous sulfur compounds coming up from the troposphere. Further, it is reasonable to assume that the continuing presence of radioactive material at this altitude is due to the influx of finely divided radioactive particles from higher elevations. That this is the case by Drevinsky and Martell (10) who have shown that the upper level debris is mainly associated with a particle radius less than 0.02μ. The Rh^{102} analysis of Kalkstein (11) (1961) also indicates the high altitude sources of debris.

Thus we can consider that the samples analyzed in this study were obtained in a region where a dynamic balance existed between the influx of radioactive material, the formation of the sulfate aerosol and the removal of both to the troposphere. If we consider that the total number of radioactive particles entering the mixing zone at approximately 20 km is small conpared to the number of ambient sulfate particles and further that the sulfate material is growing in a zone of supersaturation of a chemical precursor to this condensed sulfate, then we could consider that the radioactive particles immediately become condensation nuclei for the sulfate material. This might cause a rapid growth to the 0.01μ region, thus effectively immobilizing the radioactive particles for significant coagulative interaction. The radioactivity would therefore be distributed throughout the sulfate population approximately in proportion to the number of particles. This would produce agreement with our data since as shown in Tables 2 and 7 the impactor sample collects approximately 12% of the number of ambient sulfate particles and 10% of the radioactivity.

Further, the inclusion of the radioactive material as nuclei of the sulfate particles would produce a strong correlation between the activity of the sample and the mass of sulfate in the sample. Because of the time delay between the radioactive particle acting as a condensation nucleus and its presence in a large sulfate particle, the activity collected in an impactor sample would be biased toward particles which had entered the 20 km region sometime previously. Therefore, if this is the mechanism of interaction, one would expect fresh debris to be concentrated in the smallest particles of the sulfate distribution. It is obvious that this question could be examined by several well chosen isotopic ratio studies on samples from different stages of the cascade impactor, collected from a region containing debris from a test conducted a few months before. A further consequence of this interaction would be a strict correspondence between the activity-versus-radius distribution and the number-versus-radius distribution.

If, on the other hand, we assume that the dominant mechanism of interaction is one of coagulation, then the tendency of condensing sulfate material to grow onto the incoming radioactive particles must be weak, which would mean that either no supersaturation of the sulfate existed or that the radioactive particle surfaces are particularly inhospitable to the condensing sulfate material. In any case, the mechanics of the interaction would follow the theory outlined in Section II. We saw in this section that the activity would be attached approximately in proportion to the first moment of the number distribution and that again we would expect to collect only a small fraction of the activity on our impactor samples. However, with this mode of interaction, there would be no tendency for the fresh debris to be concentrated in the very small particles but it would more likely be distributed throughout the sulfate distribution with a maximum as indicated for the coagulative decay constant distribution at approximately 0.1μ radius.

As indicated by Junge, Chagnon and Manson (1), the residence time of the sulfate material is between six months and a year, and therefore if a large fraction of the radioactive material is to be attached to the sulfate population, the coagulation half-life of the radioactive material must be considerably shorter than this time. As seen from Table 1, particles of 0.005 μ radius and smaller would certainly satisfy this requirement. In this case there would, of course, also be a correlation between the activity collected on the impactor samples and the mass of sulfate collected.

In the case of either of the two mechanisms discussed so far, the exact distribution of activity throughout the sulfate population would depend intimately on conditions obtaining during the initial period of influx of radioactive material into the 20 km level. Since these conditions will very likely be more complicated than we have assumed, any calculations based on a uniform distribution of the debris and sulfate particles throughout a zone of the stratosphere would be expected to agree with the observed results only in rough outline.

A third explanation of our results might be that a separate population of radioactive particles exists, with average radius approximately 0.02 μ. These particles would be assumed to attach themselves to the ambient sulfate population in such a way that the particles collected by the impactor would contain 10% of the total activity, while the bulk of the activity retained its individual and separate identity. The results of Drevinsky and Martell (10), described above, clearly indicate that this is unlikely at 20 km altitude.

A series of experiments could be proposed to decide between the three interaction mechanisms described above. This would be a continuation of the simultaneous collection of filter samples and impactor samples, through the use of balloon-borne, jet impactor and filter systems similar to those used by Drevinsky and Martell, or aircraft mounted filter and impactor systems such as those used in the current work. It would be expected that the former would yield more definitive results on particle size discrimination, as well as reaching higher altitudes. However, the techniques for the analysis of non-radioactive material on these samples have not yet been worked out. This fact, together with the greater cost in money and man-hours involved in the balloon flights might dictate in favor of the aircraft system, provided that a continuing aircraft sampling program is planned.

The principle of this series of measurements would be to collect both radioactive and non-active material in a region of the stratosphere where recently created nuclear debris was in the process of mixing with the sulfate aerosol, presuming that such regions might be found. During an extended series of measurements, one would expect to find no correlation between the activity present and the sulfate material present if the radioactive particles are contained in an independent population, which in the early stages of mixing would have its concentration determined primarily by circulation patterns.

If, on the other hand, the radioactive particles act as condensation nuclei for an existing supersaturated condensing sulfate material, then we would expect the incursion of fresh radioactive debris into the 20 km level to cause an increase in the density of condensed sulfate material, and a correlation between the activity and the mass of sulfate might well be expected. We would also expect that the fresh debris would be segregated in the very small particles.

If the interaction is through the coagulation mechanism, the fresh radioactive debris would be expected to be spread throughout the sulfate aerosol population, and the proportion which would remain in a separate population at any particular time would depend on the relative rate of influx of radioactive material and the coagulation to the sulfate distribution.

The purpose of this paper is to point out that there does exist an interaction between the radioactive debris and a known naturally occurring hygroscopic material (the sulfate aerosol), and that the mechanisms of

this interaction are amenable to study by present experimental techniques. It is obvious that if a large fraction of the radioactive debris becomes attached to a hygroscopic population, the interaction of this debris with the water condensation processes taking place in the troposphere would be strongly affected. It seems likely also that the sulfur condensation cycle which apparently takes place in the lower stratosphere is effective in long range cleaning of the stratosphere of any nuclear or other debris. One might consider the sulfur cycle in the stratosphere as similar to the water condensation cycle in the troposphere, in so far as its effect on cleaning or removing otherwise long lived small particulates from this region of the atmosphere is concerned.

Our interpretation of the interaction of the radioactive debris with the ambient sulfate population has as its foundation the interpretation of the origin of these sulfate particles presented by Manson, Junge and Chagnon (5) and also by Junge and Manson (4). Whether or not this theoretical foundation is valid, the correlation between the activity and the sulfate mass presented in this paper indicates that further study of the interaction between the radioactive and non-radioactive particles in the stratosphere is required.

Acknowledgments

The author is indebted to Dr. P. J. Drevinsky and Mr. Irving H. Blifford, Jr. for their help in providing equipment, standards, and advice for the beta activity measurements. Dr. E. A. Martell and Dr. C. E. Junge have provided stimulation and support during the course of the work, in addition to many suggestions concerning the interpretation of the data.

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DISCUSSION

HOLLAND: This result is certainly interesting. It looks like it might make the radioactivity measurements or the chemical measurements more useful if one can rely on relationships of this kind. I think that the variability of each of the components is almost as interesting as the fact that the ratio is constant, and this needs to be explained also.

We will postpone the discussion of these two papers until after the next two, combining the discussions because of time limitations. (See page 145).

ATMOSPHERIC INFLUENCES ON FALLOUT DEPOSITION

PART 2. ATMOSPHERIC MOTIONS

a. Local Fallout

Session Chairman: Joshua Z. Holland, Chief
Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission

A COMPARISON OF FALLOUT MODEL PREDICTIONS WITH A CONSIDERATION OF WIND EFFECTS

Gilbert J. Ferber and Jerome L. Heffter
U.S. Weather Bureau
Washington, D. C.

Over the past decade, a great many fallout prediction models have been developed by many different organizations for a variety of uses. Most of us in the fallout "model-making" business are aware that we deal with many uncertainties which are reflected in the discrepancies between the various model predictions. However, we have not always been able to clearly define and explain these uncertainties to the users of our product. One reason for this is that the extent of the discrepancies and the reasons for them have not always been clear to us. Obviously, this is an unsatisfactory state of affairs and efforts are being made to clarify the situation. The task is not a simple one and it is more likely to lead to a better understanding of the problem than to an improvement in fallout predictions.

It may well be that the limitations imposed on us by the available fallout data make it unlikely that demonstrably better model can be devised. If this is so, perhaps our efforts should be directed, not toward the development of still more models, but rather toward a quantitative statement of the uncertainties and a study of their significance in nuclear attack casualty assessment problems.

Our knowledge of fallout is based primarily on the data collected during nuclear tests at the Nevada Test Site and the Eniwetok Proving Ground. Nuclear bursts in Nevada have been confined to yields below 75 KT. Most of these shots were detonated on towers ranging in height from 100 to 700 ft. or from balloons at heights from about 400 to 1,500 ft. Adequate fallout prediction methods have been developed for these yields and burst conditions. There have also been a few surface bursts in Nevada with yields of about 1 KT or less. Tests at the Eniwetok Proving Ground have included thermonuclear devices with yields ranging up to about 15 MT.

Most detonations took place on barges in the Eniwetok and Bikini lagoons or on the coral reefs. A few were fired on atoll islands.

In a true land surface burst over a large city, the nature of the fallout particles may be quite different from those produced in a detonation on a coral atoll or atop a steel tower. Hence, conclusions drawn from nuclear test data must be applied with caution to the rather different burst conditions likely to be encountered in a nuclear attack situation. There are also many difficulties in the analysis and interpretation of the fallout data. This is particularly true for tests at Eniwetok since the fallout descends over vast stretches of the Pacific Ocean.

Therefore, it is not surprising that the many fallout models which have been developed for nuclear attack damage assessment applications show considerable differences in the predicted fallout pattern.

Fig. 1 illustrates some fallout predictions derived from several models currently in use. The predictions are for a 1 MT, all fission, land surface burst with a constant 25 mph wind speed from the ground to the top of the cloud. The vertical wind shear*, which determines the angular spread of the fallout sector, is taken to be 0.2 knots per 1,000 ft. The contours show the hypothetical H+1 dose-rates at 3 ft. above an infinite plane. Dose rates above average terrain would be about 70% of the infinite plane dose rates.

The WSEG-RM-10 prediction is taken from the Weapons System Evaluation Group, Research Memorandum No. 10 by Pugh and Galiano.(1) This model has been used by the National Resources Evaluation Center for nuclear attack casualty assessment.

The pattern labeled WSEG-NAS is a revision of the WSEG model in accordance with the as yet unofficial and unpublished recommendations of the National Academy of Science Working Group on Fallout Models for Attack Damage Assessment. The working group was formed at the request of the Office of Civil and Defense Mobilization (now Office of Emergency Planning) to recommend a model for their use. This model represents a reasonable compromise but is certainly not intended to be the final word in fallout prediction.

The next pattern represents the Weather Bureau model(2) which was developed primarily for fallout prediction in connection with nuclear tests. The model has been revised slightly for application to surface bursts in the megaton range and has also been used in attack damage assessment exercises.

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* The vertical wind shear as used in this paper, is actually the component of the vertical wind shear, measured perpendicular to the mean wind vector. The mean wind is defined as the vector average of all the winds from the ground to some chosen level in the mushroom top of the nuclear cloud.

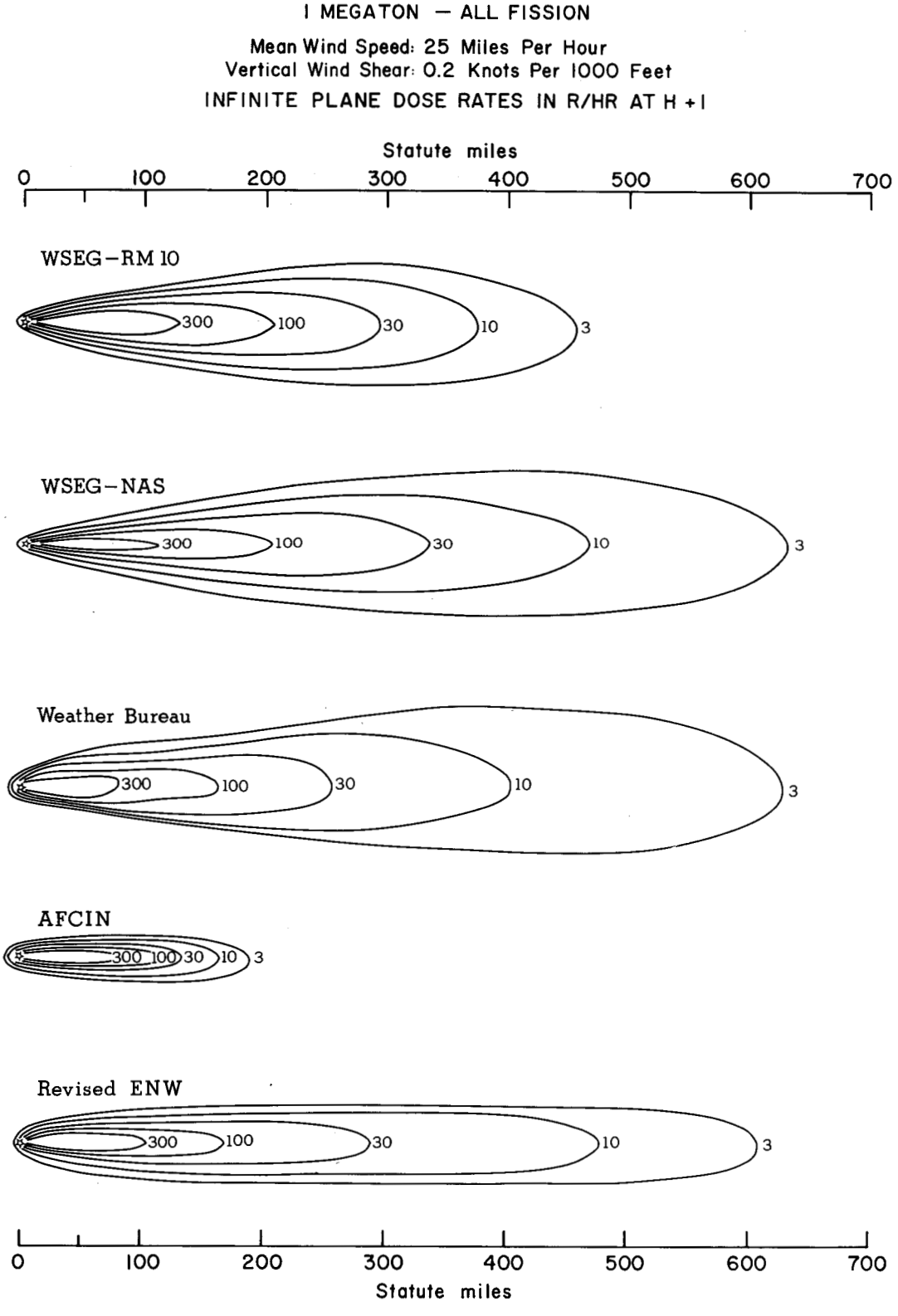

FIGURE 1. FALLOUT PATTERN COMPARISONS - 1 MT.

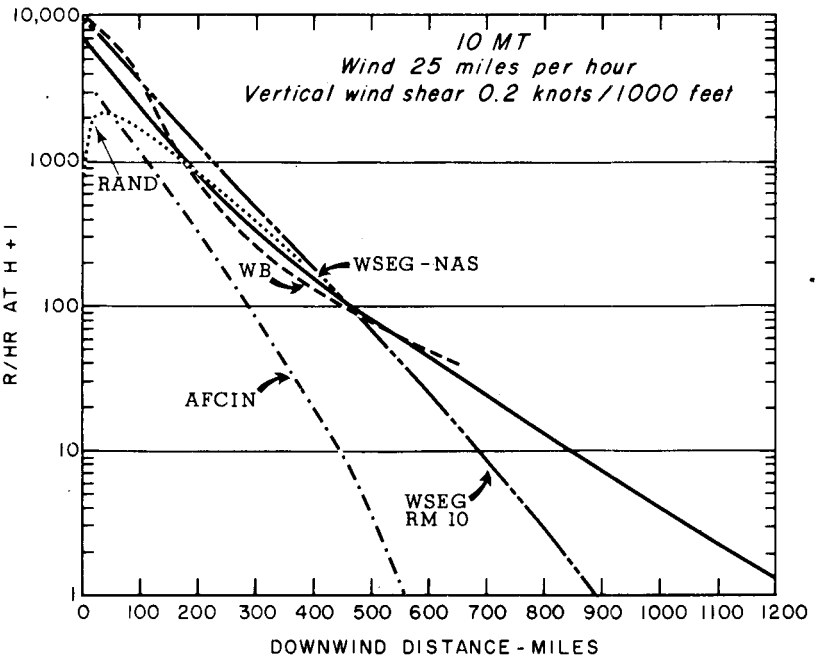

FIGURE 2. DOSE RATE - DISTANCE CURVES - 1 MT.

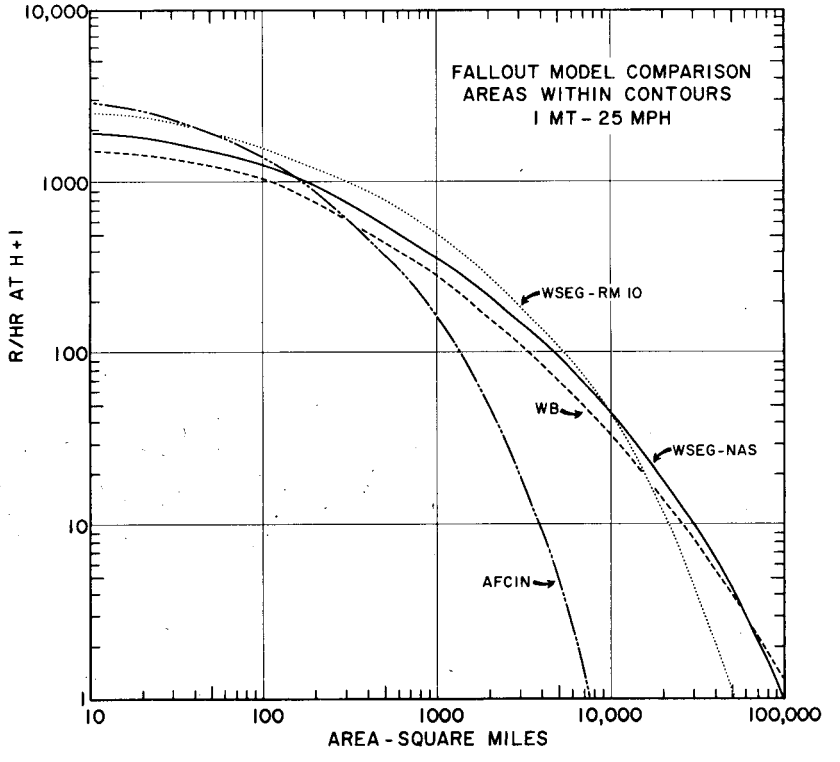

FIGURE 3. DOSE RATE - AREA CURVES - 1 MT.

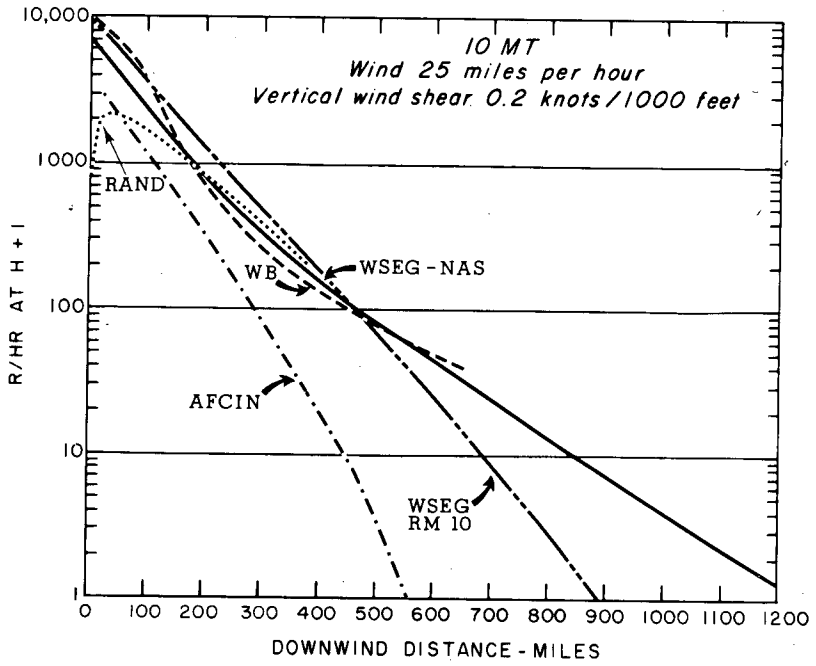

FIGURE 4. DOSE RATE - DISTANCE CURVES - 10 MT.

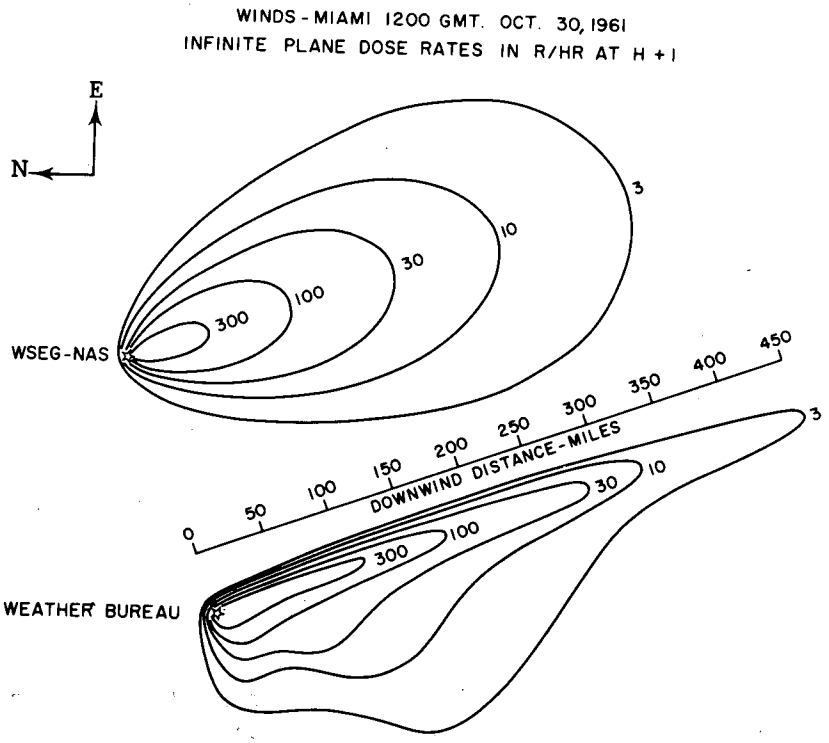

FIGURE 5. COMPARISON OF IDEALIZED PATTERN AND DETAILED COMPUTATION - 1 MT.

The AFCIN model[3] was developed by the Air Force Intelligence Center primarily for the assessment of the effects of a nuclear counterblow on enemy territory. The last pattern is one which will appear in the new edition of The Effects of Nuclear Weapons.[4] It is based on the Weather Bureau model with simplified scaling laws which enable the user to adjust for weapon size or wind speed.

At first glance, the most striking feature is the disparity between the AFCIN prediction and all the others. One may wonder about the outcome of a war game in which the AFCIN model is used to determine the fallout situation over enemy territory while our own fallout problem is evaluated with one of the other models. Actually this picture is somewhat misleading in that the greatest differences among the models appear in the lower dose-rate contours, well below the casualty-producing range.

In Fig. 2, we have plotted the hypothetical H+1 dose rate as a function of the distance from ground zero for some of the patterns shown in Fig. 1. Note that the extent of the 1 r/hr. contour varies from 200 to 800 mi. with different models but the uncertainty in the prediction decreases for the higher dose rates. Also notice that the AFCIN model predicts higher dose rates than the other models in the first 50 mi. A portion of the curve predicted by the Rand Corporation model[5] is also shown. The Rand model is interesting in that it predicts a lower peak dose and the peak occurs at some distance from the burst point.

In Fig. 3 we are still looking at the same predictions. Here we have plotted curves of H+1 dose rate versus area covered, for several of the models. The areas under the curves represent the total activity in the fallout patterns within the 1 r/hr. contour. The total activity is 2,500 r/hr. mi.^{2}/KT in the WSEG-RM-10 pattern, 2,400 r/hr. mi.^{2}/KT in the WSEG-NAS pattern, about 2,000 r/hr. mi.^{2}/KT in the Weather Bureau pattern and about 800 r/hr. mi.^{2}/KT in the AFCIN pattern. Thus, there is approximately a factor of 3 difference in total activity within the 1 r/hr. contour as predicted by AFCIN and WSEG-NAS. The activity total in the WSEG-NAS model is that recommended by the Academy of Science Working Group on Fallout Models as a best estimate. However, the number is still open to question and there is no clear proof that the AFCIN total is incorrect.

Note that the models differ not only in the total amount of activity which appears within the 1 r/hr. contour but also in the distribution of the activity. The AFCIN model puts even more activity in the very high dose-rate contours than do the other models. Thus multiplying the total activity by a factor of 3 would not bring AFCIN into line with WSEG-NAS, if the assumed activity distributions remain unchanged.

Fig. 4 shows the predicted dose rate versus downwind distance for a 10 MT, all fission, surface burst with the same wind conditions as before. For this yield, the extent of the predicted 1 r/hr. contour varies from 550 to over 1,200 mi. downwind. The 1,000 r/hr. predictions vary from 100 to about 230 mi. It is also interesting to look at the various dose-rate

predictions at a given distance. At 100 mi., for example, the predicted H+1 dose rates vary from 1,000 r/hr. to 3,500 r/hr. At 300 mi. the range is from 80 to 500 r/hr.; and at 500 mi., from 3 to 80 r/hr.

Notice that the AFCIN curve now falls entirely below the other curves, except for the Rand prediction in the first 50 mi. Thus, we see that the relationship between model predictions is not constant, but varies with the size of the detonation. It is this sort of thing which makes it impossible to reconcile the various models without delving into the details that went into their development.

So far, we have compared idealized, cigar-shaped patterns under uniform wind conditions. In Fig. 5, we have used an actual wind sounding with about 90° of angular shear to compare the WSEG-NAS idealized pattern with resulting from a detailed computation with the Weather Bureau model. The WSEG patterns are superior to most idealized patterns in that they incorporate vertical wind shear as a variable in addition to wind speed. Due to the large angular shear and somewhat lower wind speed, the patterns are shorter and wider than those in Fig. 1. However, the idealized pattern is symetrical about an axis which lies along the direction of the mean wind from the ground to the lower portion of the mushroom head of the nuclear cloud. The detailed computation, which takes into account the winds at all levels, results in an asymetrical pattern. The "hot-line" is in the same direction but there is no fallout to the east of this line and a considerable bulge to the west. It is evident that the idealized pattern, based on a single mean wind, does not always present an accurate picture of the fallout area.

In Fig. 6 we turn for a moment to fallout prediction for the critical area immediately upwind of the ground zero. Since the mushroom cloud from a 10 MT detonation has a radius of about 27 mi. (at 10 minutes after burst), fallout might be expected to occur at a considerable distance upwind of the ground zero. The upwind fallout estimate from the "Capabilities of Atomic Weapons", Technical Manual 23-200[6], which is shown here, is typical of the earlier estimates of upwind fallout based on Pacific test data. The curve is labelled "Old TM 23-200" since a revision of this manual is in preparation. With a 25 mph mean wind the 10 r/hr. line was predicted to extend about 26 miles upwind. More recent studies, here represented by the Weather Bureau[7] and WSEG-NAS predictions, indicate that the upwind fallout problem will probably be less severe than was formerly believed. The reason for this is that the Pacific tests were conducted under conditions of very light mean winds, generally less than 10 mph. The mean winds in the United States are much stronger, generally over 25 mph and, in winter often exceeding 60 mph. Fallout particles from the upwind portion of the nuclear cloud would be carried back toward ground zero by these winds, thus decreasing the upwind extent of the fallout pattern. The Weather Bureau and WSEG models predict that the 100 r/hr. contour will extend 4 to 7 mi. upwind rather than 11 mi. The 10 r/hr. line is predicted at 7 to 10 mi. rather than 26 mi. and virtually no fallout is expected beyond about 12 mi. upwind. This is for a

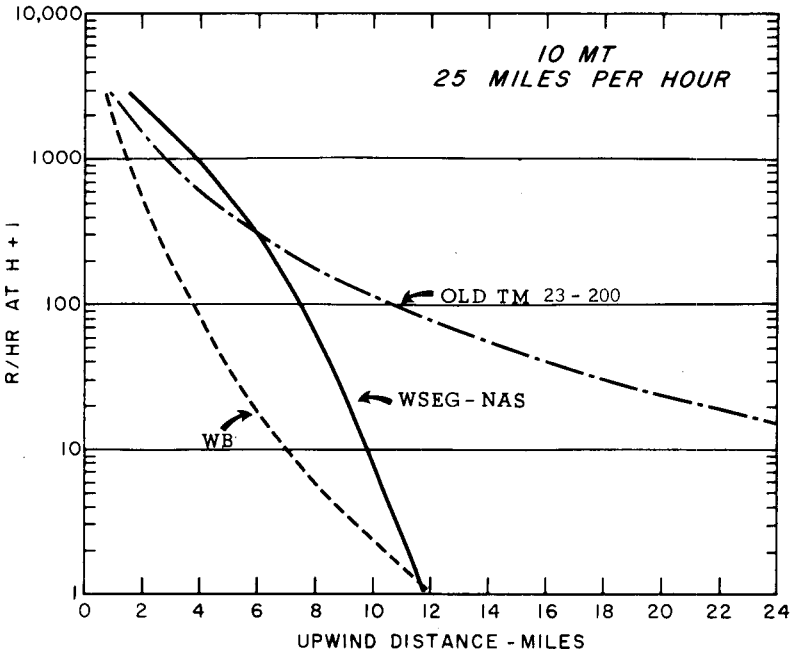

FIGURE 6. UPWIND FALLOUT PREDICTIONS.

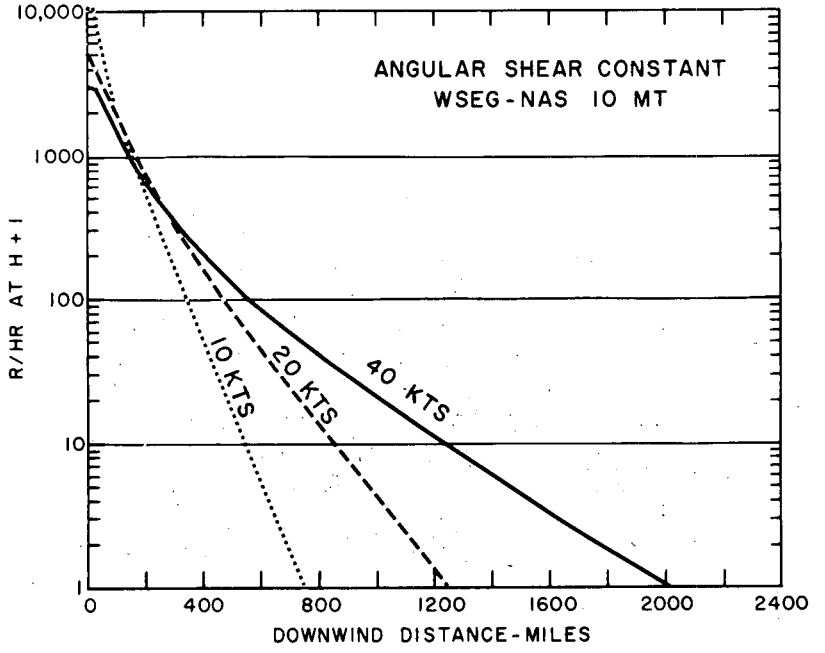

FIGURE 7. EFFECT OF WIND SPEED.

25 mph mean wind from the ground to about 40,000 ft. With higher winds there would be even less upwind fallout. The predictions for the very high dose rates have not changed significantly but these are rather academic since they are within the area of nearly total destruction from blast and fire.

Next we will take a look at the effects of wind speed on the downwind fallout pattern. In Fig. 7 curves of H+1 dose rate versus downwind distance are shown for the WSEG-NAS 10 MT patterns for 10, 20 and 40 knot mean winds with the same angular wind shear in all cases. Note that increasing the wind speed lowers the dose rates close to ground zero and increases the dose rates at greater distances. The dose rate at a given distance may vary by more than a factor of 10 due to wind speed alone. At 600 mi., for example, the dose rate varies from 5 r/hr. with a 10 knot wind to 45 r/hr. at 20 knots and 90 r/hr. at 40 knots.

The effect of wind speed can be described by a very simple scaling law. If the angular spread is kept constant, doubling the wind speed will result in $\frac{1}{4}$ the dose rate at twice the distance. Thus, with a 10 knot wind we have 4 r/hr. at about 625 mi. With a 20 knot wind we have $\frac{1}{4}$ the dose rate, or 1 r/hr. at twice the distance, or 1,250 mi.

Fig. 8 shows the effect of varying the vertical wind shear which governs the angular spread of the fallout sector. Again we have curves of dose rate versus downwind distance for a 10 MT burst with 20 knot mean winds. In going from 0.1 to 0.2 knots/1,000 ft. we are doubling the angular spread, and doubling again in going from 0.2 to 0.4 knots/1,000 ft. In general the dose rate at any distance is inversely proportional to the angular spread. At 380 mi., for example, the dose rate is 400 r/hr. at H+1 with a shear of 0.1 knots/1,000 ft. Doubling the shear, we obtain $\frac{1}{2}$ the dose rate, 200 r/hr.; doubling again, we obtain 100 r/hr. Very close to ground zero the change is much smaller since the diameter of the nuclear cloud is large compared to the spread due to directional wind shear. In general, the angular wind shear decreases with increasing wind speed, so that strong winds will tend to deposit the fallout in long narrow patterns, with high dose rates extending out to great distances.

We will now focus on predictions in the critical area where the radiation dose is in the lethal range. In Fig. 9 we have drawn contours of the cumulative 4-day dose to a completely unsheltered population. The patterns are for a 10 MT fission yield with a 25 mph mean wind. A factor of 0.7 was used for terrain shielding but no other shielding or biological repair factor was included in the calculation of the 4-day dose.

It is estimated[4] that no fatalities will occur with an exposure of less than 200 r, 450 r will result in about 50% fatalities and exposure to about 700 r or more over a period of a few days will result in close to 100% fatality.

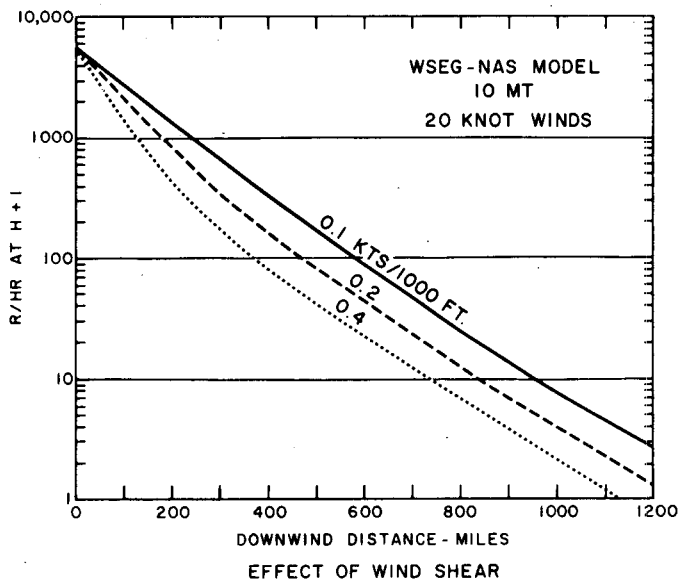

FIGURE 8. EFFECT OF WIND SHEAR.

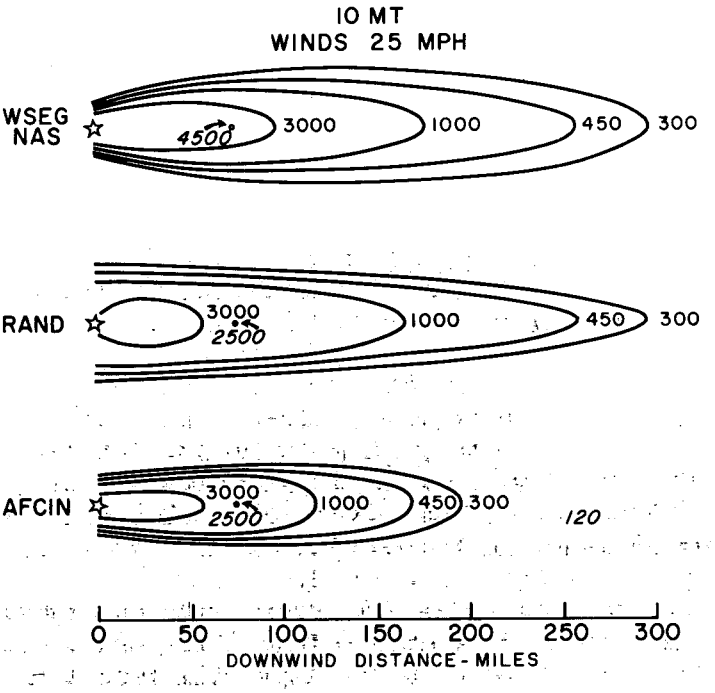

FIGURE 9. COMPARISON OF 4-DAY (ROENTGENS) PATTERNS - 10 MT.

Both the WSEG-NAS and Rand models predict 100% fatalities for an unsheltered population in an area about 40 miles wide extending about 200 mi. downwind. In the AFCIN prediction, this area extends only 150 mi. downwind. At a distance of 260 mi., where WSEG-NAS and Rand predict 450 r or 50% fatality, AFCIN predicts 120 r, or no fatalities.

Now if we assume a sheltered population, with an arbitrary shielding factor of 10, an outside dose of 4,500 r is required to produce 50% fatalities. A shielding factor of 10, or better, is provided by the basement of a frame house. The WSEG model predicts 4,500 r at 75 mi. while Rand and AFCIN are now in agreement with only 2,500 r at this distance. The area in which WSEG and AFCIN would predict 100% fatalities (7,000 r or greater), even with a shielding factor of 10, is about 10 to 20 mi. wide and, at most 60 mi. long. However, according to the Rand model, an outside dose of greater than 7,000 r may not occur anywhere in the fallout pattern for this yield and wind speed.

Fig. 10 shows the effect of wind speed on casualty estimates based on the WSEG-NAS prediction for a 10 MT fission yield. The hatched areas indicate, for wind speeds of 10, 20, 40, and 60 knots, the percentage of deaths due to fallout which is expected along the axis of the fallout pattern as a function of distance from ground zero. In computing the doses for the various wind speeds it was assumed that the angular wind shear is inversely proportional to the wind speed. The widths of the hatched areas reflect the uncertainty in the relationship between dose and biological effect.

First, consider the case where the population has no shielding whatsoever. With 10 knot winds, we may expect 100% fatalities out to about 140 mi. with no fatalities beyond about 200 mi. With a 60 knot wind, 100% fatality is expected out to about 400 mi. with no deaths due to fallout beyond 700 mi. Wind speed is evidently a critical factor here. However, when we apply a shielding factor of 10 to the same dose predictions, the results are very different. The area of 100% fatality extends about 50 mi. regardless of the winds. With a 10 knot wind, no deaths are expected beyond 90 mi. and even with a 60 knot wind there should be no fatalities beyond 220 mi. The effect of shielding is quite striking.

Summary

If we are interested in the entire fallout pattern produced by a single nuclear burst, we find that the various models can give very different results. Also, wind conditions can drastically effect the dose-distance relationship as well as the orientation and shape of the pattern. If our interest is confined to doses in the lethal range, and especially if a shielding factor is added, we find that the discrepancies among the various model predictions are smaller and the effect of wind speed on these high doses is also much smaller. Nevertheless, these differences are still quite important in the critical range from about 200 to 700 r where relatively small changes in dose result in large changes in the fatality estimates.

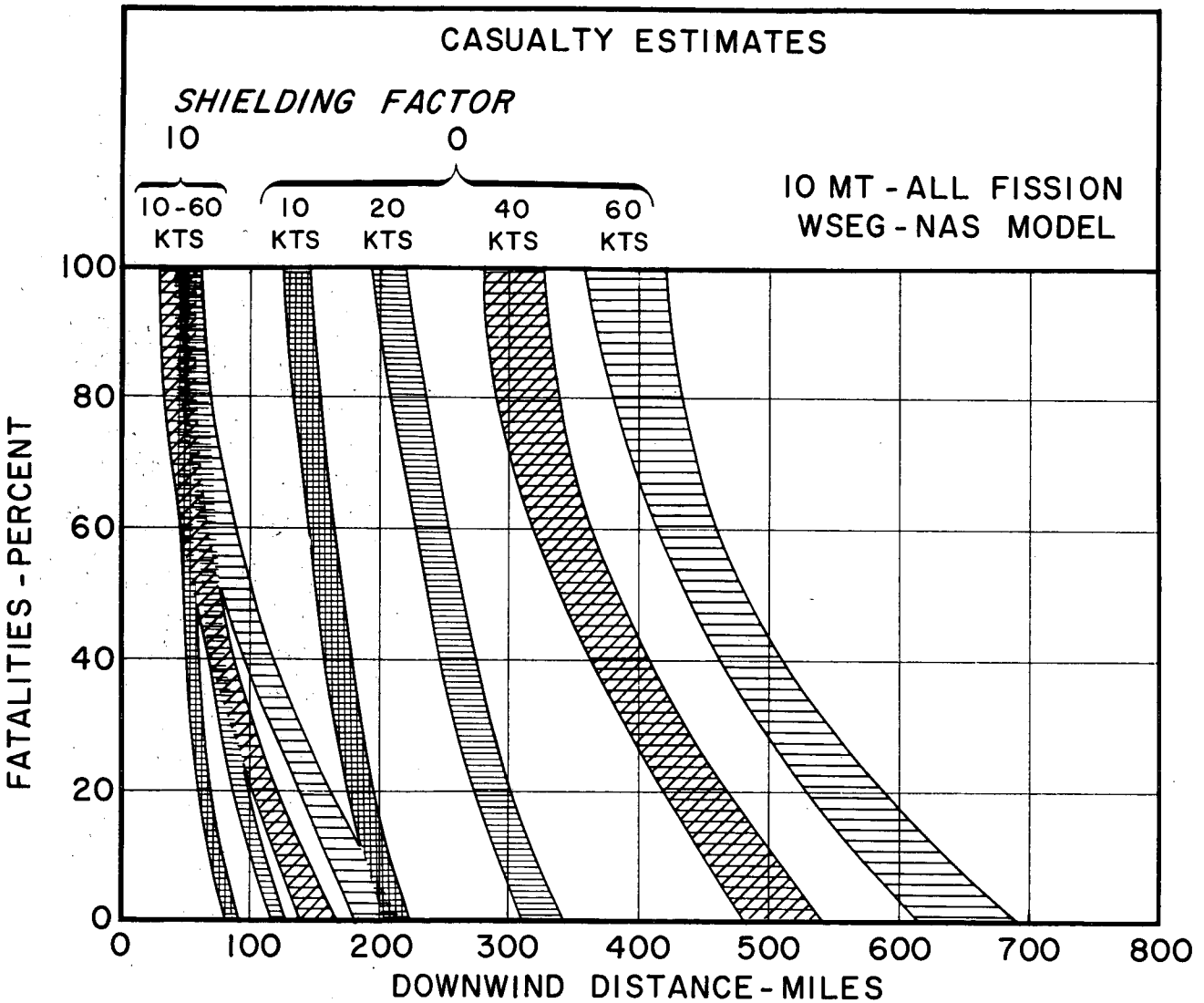

FIGURE 10. EFFECT OF WIND SPPED AND SHIELDING ON CASUALTY ESTIMATES - 10 MT.

Finally, when we consider nuclear attack damage assessment on a national scale, with the additive effects of hundreds of nuclear bursts, the significance of the fallout model uncertainties and wind conditions for the over-all casualty estimate is not at all clear. With the over-lapping of fallout patterns from many bursts, the lower dose-rate contours, for which predictions are least consistent may become important. It might prove quite interesting and enlightening to run a damage assessment problem several times for a selected attack situation, using a different fallout model each time. Then the uncertainty due to the fallout model may be evaluated and compared, for example, with the uncertainty in the shielding assumptions. In this way, the accuracy and precision required of fallout models for attack damage assessment can be determined in a realistic manner.

Similarly, the effect of wind forecast errors can be evaluated by running problems with both forecast and actual winds. The results of past damage assessment exercises seem to suggest that wind conditions do not greatly affect the total number of casualties. However, the winds will determine where the fallout casualties will occur.

We must learn to live with the fact that large uncertainties are inherent in all fallout model predictions. Further, there is little reason to hope for significant improvements in the absence of a series of nuclear tests designed primarily for the scientific study of fallout phenomena. Hence, error analyses and the development of statistical techniques to handle the uncertainties are at least as important as continued efforts to improve the fallout models for nuclear attack damage assessment.

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THE EARLY TRANSPORT OF NUCLEAR DEBRIS

Phillip W. Allen, Frank D. Cluff and Isaac van der Hoven
Research Station
U. S. Weather Bureau
Las Vegas, Nevada

Introduction

The announced purpose of this conference was to provide a review of new information and developments in fallout research, and to discuss future needs of the program. I will discuss a few relatively minor improvements in our ability to predict close-in fallout, and some of the needs of the purely meteorological end of fallout prediction. My concept of these needs comes from my observation and study of the activities of United States weather and fallout prediction units since Operation SANDSTONE in the Pacific in 1948, and more particularly from the work of the Weather Bureau Research Station at the Nevada Test Site since 1956. This station has served as the Weather Prediction Unit for all continental United States nuclear tests since Operation PLUMBBOB in 1957 and is continuing in this capacity for the tests of nuclear rocket and ramjet engines, which produce very low levels of atmospheric contamination. If, in my remarks, I use the term nuclear debris it is because the word "fallout" implies particles, and this is only part of the problem. The gaseous components of nuclear debris also present an environmental problem, particularly in reactor tests.

Variability

Outside of that posed by the mere existence of debris or fallout, the

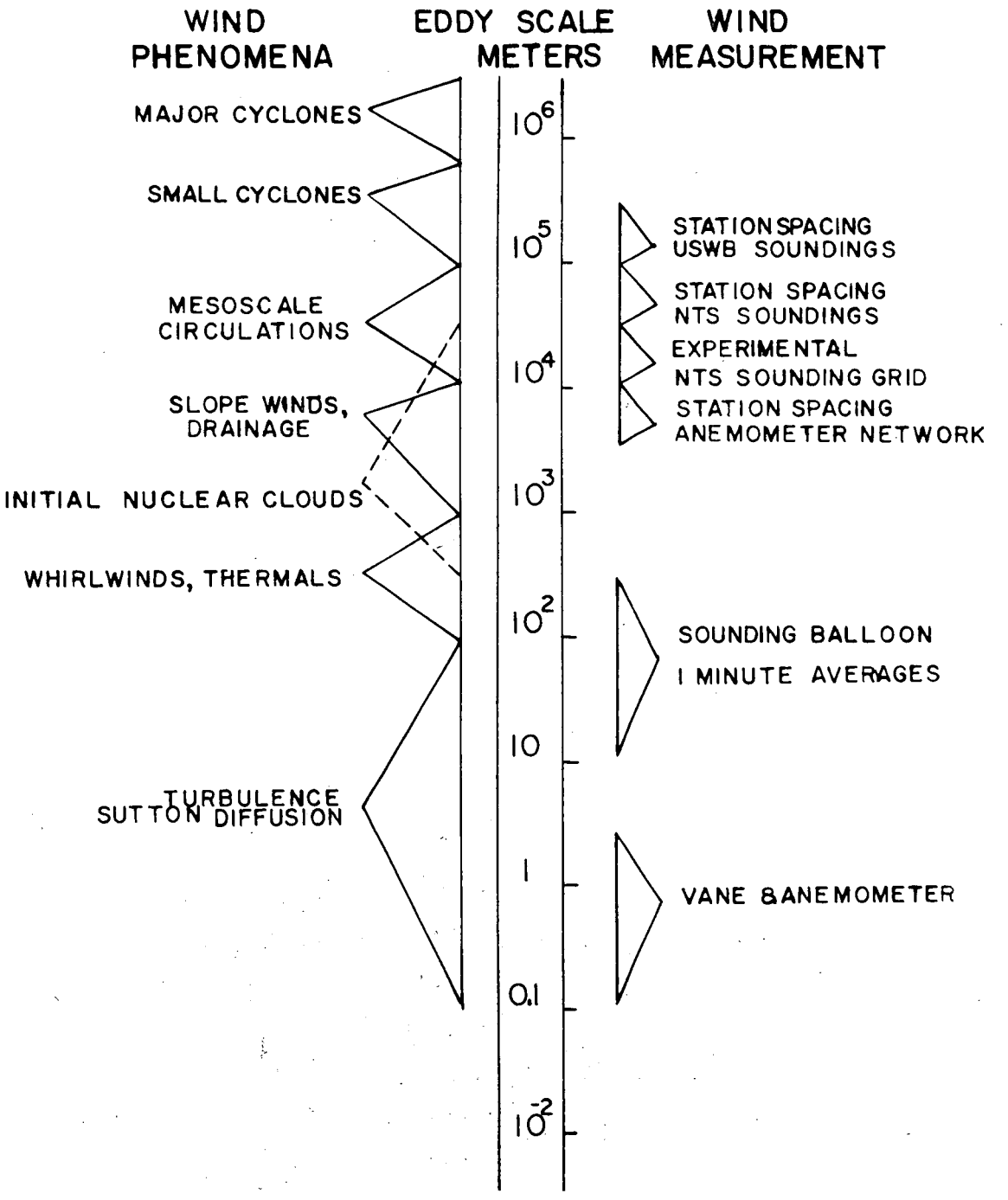

FIGURE 1. SCHEMATIC DIAGRAM OF THE SPECTRUM OF ATMOSPHERIC MOTIONS AND METHODS OF MEASUREMENT.

one problem that is the most difficult to solve, the one which gives operations people greatest concern, and which results in the otherwise needless cost of many thousands of dollars for some nuclear events, is wind variability. This characteristic of air motion has defied prediction, and in most meteorological considerations has been taken as one of the facts of life, something we just live with and accept. However, in this field where the transport of radioactive debris is involved, perhaps closer investigation is worthwhile.

A schematic of the spectrum of atmospheric motions is shown in Fig. 1. The sizes of eddies or the lengths of wave motions range all the way from Brownian motion on the small end to hemispheric on the large end. All scales exist every day, but the frequency of occurrence of a given scale may change from day to day or from place to place depending on the existence or suppression of the appropriate driving mechanism. Not all scales of eddies are driven in exactly the same way but most of them are influenced by the distribution of pressure and heat in the atmosphere, and this changes with season, cloudiness, surface slope, etc.

The size range of nuclear clouds, at time of stabilization following explosions, falls roughly midway in this spectrum, being larger than whirlwinds but smaller than such mesoscale circulations as thunderstorm high pressure cells. They cover about the same horizontal areas as drainage winds in a mountainous area but have must greater vertical extent.

On the other side of Fig. 1 are shown the scales of measurement of winds. At the low end the average wind vane and anemometer respond to circulations of the order of a meter or more across. A rising sounding balloon, on which positions are plotted every minute, will indicate motions of the order of tens or hundreds of meters across, and has the advantage of measuring at high altitude. No single instrument or device does a very complete job of recording motions larger than this, although rocketsondes with chaff clouds and smoke puffs are used effectively at very high altitudes, and constant level balloons are useful at individual altitudes.

To describe completely the motion of an eddy in any scale it is necessary to sample frequently or continuously at several points in the eddy. The spacing of the Weather Bureau's upper air sounding stations is between 10^5 and 10^6 meters (60 to 300 miles). This is adequate to define cyclones and anticyclones, upper level ridges and troughs of pressure systems that carry major changes of weather. However, mesoscale circulations are frequently not detected or if detected are poorly defined by this network.

For this reason, with the testing of nuclear weapons it became necessary to have upper air soundings closer together. The usual Nevada Test Site (NTS) sounding network for large atmospheric tests has had a station spacing of about 50 miles. In recent years with nuclear rocket engine

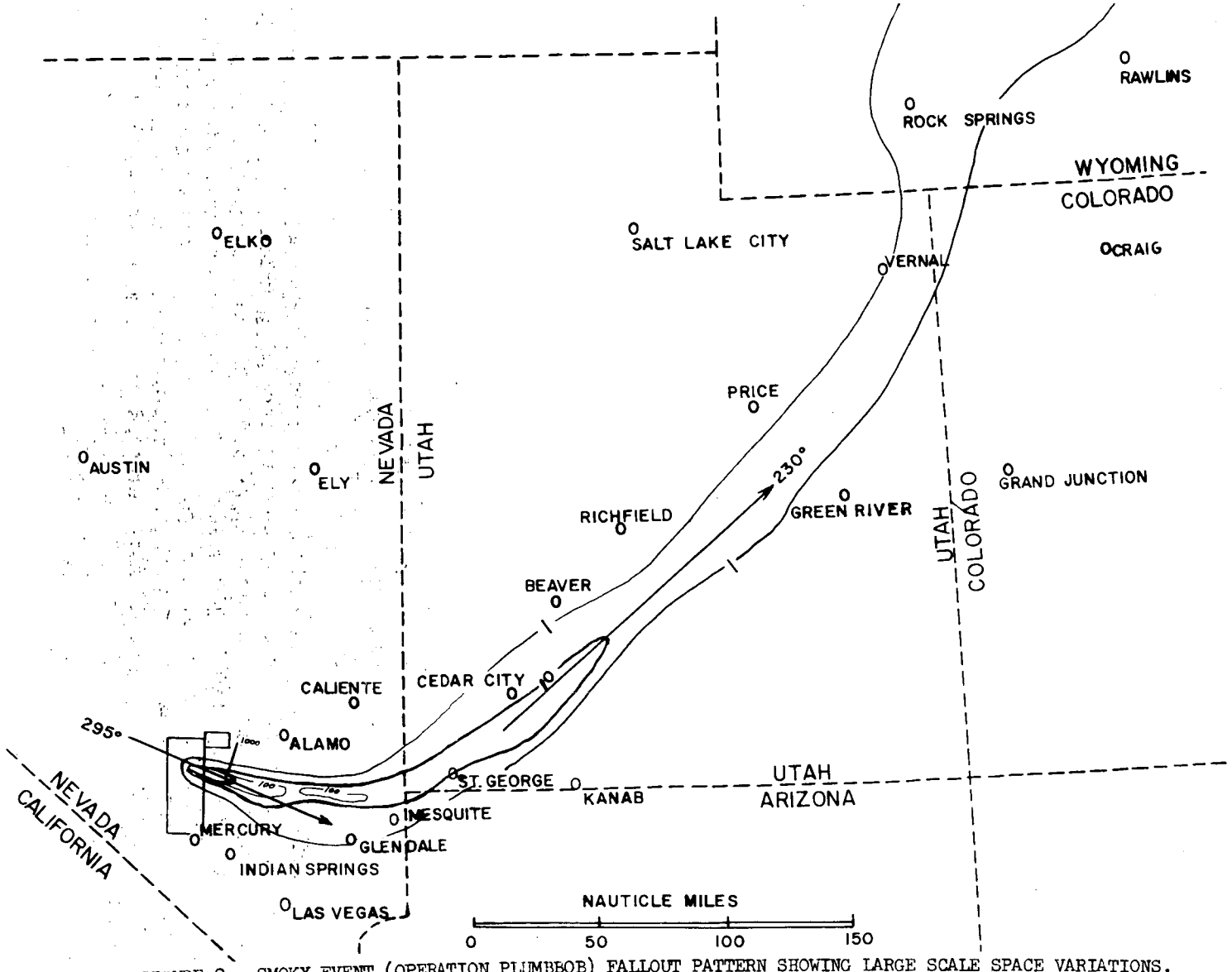

FIGURE 2. SMOKY EVENT (OPERATION PLUMBBOB) FALLOUT PATTERN SHOWING LARGE SCALE SPACE VARIATIONS.

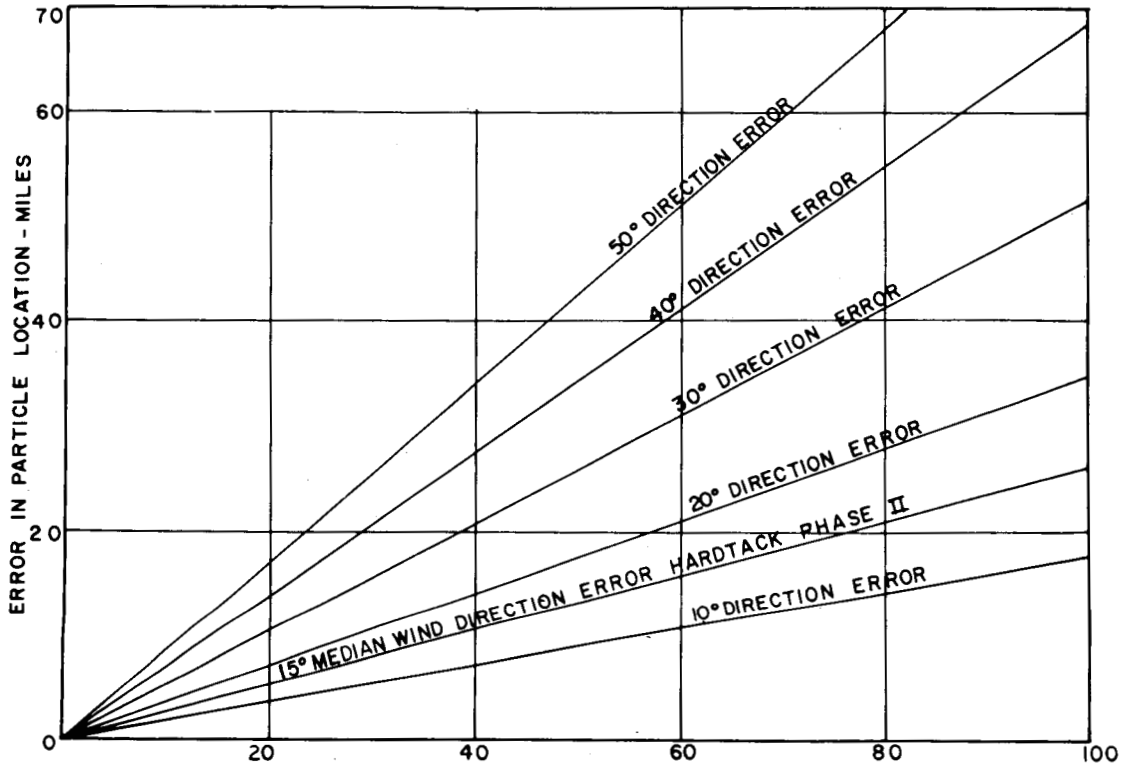

FIGURE 3. DISTANCE FROM GROUND ZERO-MILES.

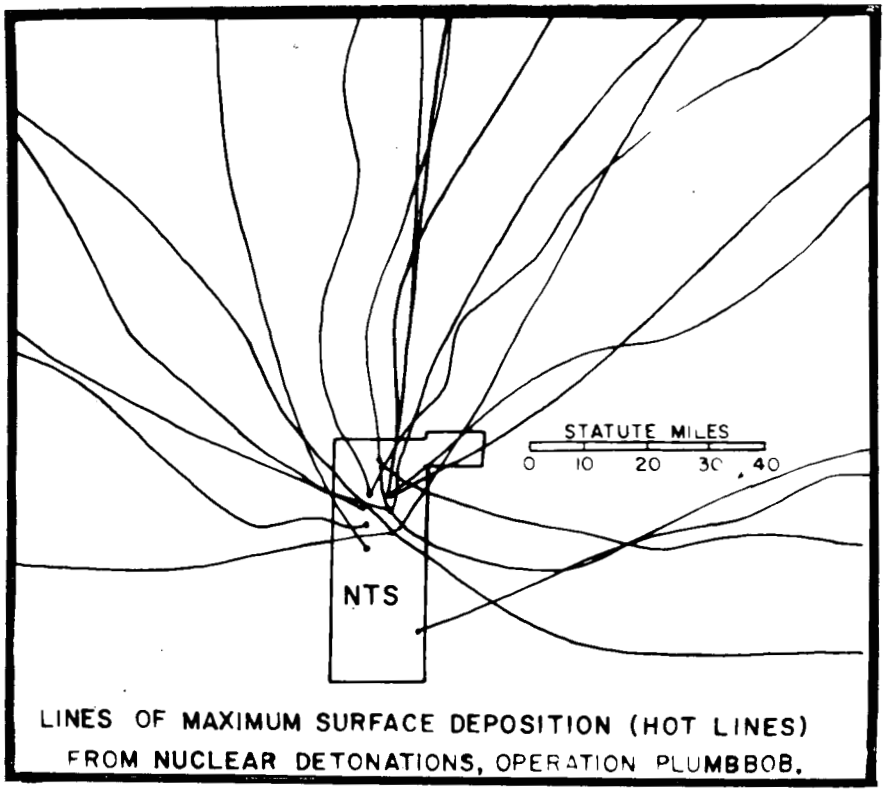

FIGURE 4.

tests it has become necessary to reduce the spacing to something like 15 miles between stations. With three or four such stations it is possible to detect most waves or perturbations of a size that would carry a small radioactive cloud astray. Anemometers on towers are located in an even denser grid to detect smaller surface eddies and slope-induced winds. Much of the meteorological study in progress at the NTS during recent years has been directed toward a full understanding of slope winds and mesoscale circulations peculiar to the area of southern Nevada. Other work has the objective of handling predictions statistically, in which the probability of occurrence of a given wind is made available. These studies have resulted in a definite improvement in the weather service provided for the Nevada Test Organization.

Why are we so concerned about these relatively small scale wind perturbations? For two reasons, one being that small perturbations make individual wind soundings non-representative, thereby possibly influencing the forecaster to make a wrong decision, and the other because the presence of a perturbation at zero time may well result in a nuclear cloud being carried in an unpredicted and undesirable direction.

The fallout pattern from the Smoky event in Operation PLUMBBOB, shown in Fig. 2, illustrates how the best fallout prediction in the world can be of little value unless the wind directions and wind changes are predicted correctly. Long narrow patterns like this would fall on very little of the predicted area if the wind direction forecast were off by as much as 10°.

Fig. 3 shows how far from a predicted location fallout particles will strike the ground for each 10° interval of wind direction error. The prediction group for Operation HARDTACK II has a median error of 15° in 6-hour predicted wind directions between 15,000 and 25,000 ft. At a distance from the Site of 40 miles this represents an error of 10 miles in cloud location. Several direction errors were as high as 30° or 40°. Fortunately, none of these errors involved significant radiation fields with respect to people. This safety feature was not accidental because in these cases it was known in advance that the directions might vary greatly, and speeds were low enough that heavy fallout would not reach inhabited areas. These factors were considered in decisions to proceed with the tests.

Fig. 4 shows the "hot-lines" or center-lines of maximum fallout for the Operation PLUMBBOB events. This illustrates the effects of perturbations and local circulations on fallout patterns. The wave lengths of these perturbations are from roughly twenty miles to values larger than the area shown, over 150 miles across. Smaller perturbations undoubtedly escaped detection in the analysis.

We have no very good information on the life expectancy or period of persistence of individual mesoscale waves but several have been observed to persist for periods of half an hour or more whereas only a few

of the larger ones have been traced through a 6-hour period in Nevada. It is completely impossible, with networks, sounding procedures, and data handling facilities now in use, to detect, measure and predict or extrapolate perturbations of this scale. We can detect them and sometimes can indicate roughly their maximum effect on a cloud trajectory. Sounding balloons take from several minutes to an hour to rise through the layers in question and several more minutes are required to process the data. By the time this has occurred and the forecaster can evaluate the meaning of the data and compute future motion the wave will have moved through the area or will have disappeared. A good example of this occurred with the test of the reactor Kiwi A in 1959 (Fig. 5) when a perturbation of at least an hour duration caused the motion of the cloud of the exhaust to depart from the predicted 190° wind direction by 15° on one side then return to 10° on the other side. The activity level for this event was very low and this departure is only of interest as an illustration of what can happen.

Many of the eddy motions in the atmosphere must now be considered to be random in the sense that their cause, source, history and dimensions are obscure. The speed and accuracy of the NTS radar wind plotter and surface weather network have contributed much to operational fallout safety. However, we believe that it will eventually be within our capability to handle intelligently, operationally, and on a dynamic rather than statistical basis, smaller scales of motion than we now do, and we suggest that more of our limited meteorological talent might well be applied in this direction.

Future Needs

The obvious goal in all air polluting activities should be, and is, to eliminate sources of pollution. However, the simple fact remains that the number of sources of radioactive and toxic pollutants is growing steadily. Instead of wishing them to hurry up and go away, meteorologists should be preparing to live with some of them for a long time.

The ultimate goal in meteorological support of projects involving toxic air pollution, whether from A-bomb fallout, nuclear engine exhaust gases, toxic chemical rocket exhausts, or industrial waste, should be, in my opinion, to develop a high speed weather data handling and computing system for mesoscale application, to go along with the development of a large-scale forecasting system of the National Meteorological Center's Numerical Unit. This mesoscale unit should include:

1. A close network of stations around each source, reporting upper air data frequently or even continuously. Network grid spacing horizontally and vertically should be designed to detect wind pressure and temperature changes which result in changes of wind and

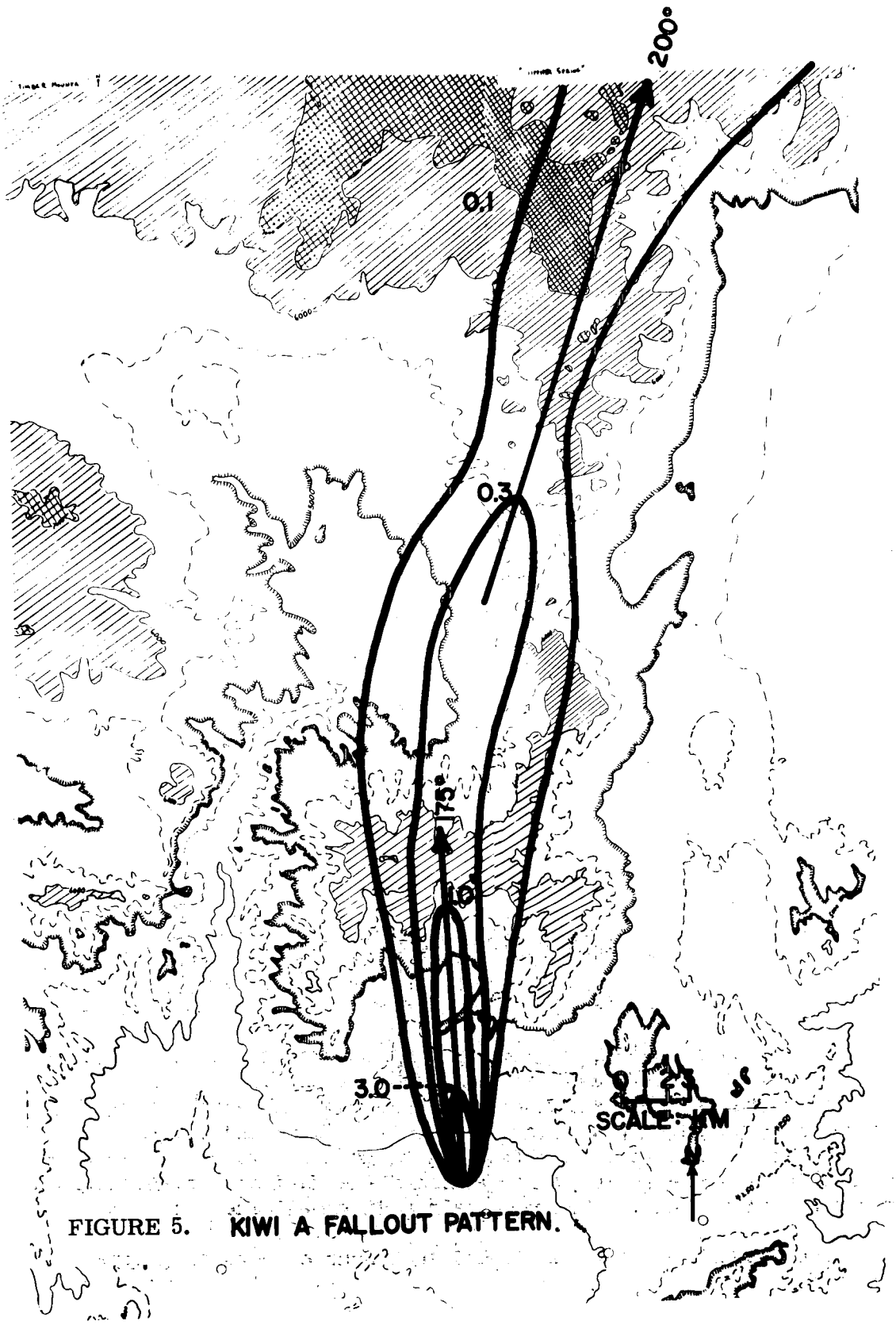

FIGURE 5. KIWI A FALLOUT PATTERN.

stability, just to determine the appropriate grid spacing requires knowledge not now available. Some development is already being made of means for continuously reporting upper air data.

2. Instantaneous telemetering of information to central analysis and computing facilities. This is now physically possible, but very expensive.

3. Computing stream functions and temperature field over the grid, then extrapolating these perhaps as much as 6 hours into the future. The real obstacle to this system is the development on mathematical models of mesoscale processes, and this is where additional research should be concentrated.

4. Distribution of forecasts to users. Such a high speed system would be of little value operationally unless its results were available and useful immediately. Specific applications should include forecast interpretation to the extent that acceptable forecasts would be so labeled and those presenting weather unacceptable for the particular operation would be clearly marked to eliminate loss of time for forecast interpretation on the part of the operations people.

Whether the large amount of effort necessary to realize such a complete mesoscale prediction system would be worth the cost in value to weapons test fallout prediction alone is difficult to determine. It would require investing effort in research units that are better equipped for this kind of work than our small test support group in Nevada. We are in the position that many close-in fallout patterns have been laid down without obvious change in the environment of the Nevada Test Site, and without excessive exposure of the off-site population to fallout radiation. This has been accomplished simply by waiting until atmospheric patterns of motion occurred in which it was possible to assure fallout safety. But these delays are costly and it might be possible to reduce them somewhat with more effective and confident use of marginal weather situations. The primary justification for this system in Atomic Energy Commission operations would probably be in its contribution to safety through establishing rapid and accurate indications of wind both currently and for a short time into the future.

DISCUSSION

Mr. Joshua Z. Holland, Chairman of these two sessions, directed the discussions of the papers of the sessions which follows.

HOLLAND: Thank you very much. I don't know whether you realize how hard it was to get Phil (ALLEN) to come or for Phil to arrange to come to this conference under the circumstances that exist in Nevada these days. We are very grateful for the efforts that you have made in preparing this paper.

Next, we will have a discussion of Mr. FERBER'S paper, Mr. ALLEN'S paper and Mr. MANSON'S paper.

MACHTA: May I ask whether the radioactivity which you (MANSON) found in the stratosphere, the gross beta activity was necessarily fission products, or could it have been cosmic ray-produced, or other natural radioactivity?

MANSON: This was very probably due to fission products since the samples were counted a week ago and collected between a year and several months previously. The measured apparent half-life and absorption curve of this activity is also consistent with fission products, which are known to make up the major part of the activity at 20 km altitude.

Walter C. CONOVER (U.S.Army Signal Research and Development Laboratory, Ft. Mommouth, N.J.): I have a question to direct to Mr. ALLEN. He is concerned with the lack of measurement of mesoscale winds. I would like to know if a test produces a pertubation in the mesoscale wind field, thus invalidating the prior wind measurements that are made? Does he have any knowledge on this subject?

ALLEN: We have reason to believe that almost microscale changes in the circulation pattern do occur. By this I mean very near the surface and within fractions of a mile of the test or the shot. But we believe that beyond the distance of about a mile in the case of low-yield events, or a very few miles in the case of large-yield events there is no change in the meteorological system of any measurable significance.

FRIEDLANDER: My question is directed to MANSON. We have been very interested in the regularities that you found in the size spectra of the stratospheric aerosol which would tie in with the irregularities observed in the troposphere. Would you care to comment on the relationships? You mentioned before that you felt that your later data indicate that there has been some change with respect to the shape of the spectrum in the stratosphere. Is it now closer to the distribution that has been observed in the troposphere?

MANSON: In the troposphere $\frac{dv}{d(\log r)}$ is constant between about a tenth of a micron and larger, one micron, I believe. The concentration or the number distribution in the stratosphere, I believe, has the same variation with radius as in the troposphere in this size range, but the concentration is a factor of 10^4 lower.

The implication, of course, is that possibly this is material just mixed from the troposphere, and we have a layer of tropospheric aerosol existing just above the tropopause. But the chemical and physical characteristics of the particles in this layer are extremely uniform, and we still feel that the stratospheric material is not of direct tropospheric origin, as a condensed phase, but is converted in the stratosphere to the condensed phase by oxidation of SO_2. I really have no explanation for why the two size distributions should be the same. This is, inverse with the cube of the radius, $\left(\frac{dn}{d(\log r)} = Cr^{-3}\right)$.

Edwin F. DANIELSEN (University of Washington, Seattle): I direct my question to MANSON. What was the counting date of the three samples for which you gave the ratio of beta activity to mass?

MANSON: The tip samples were counted in October 1961 and the filter samples were counted in November 1961, separated by approximately a week. But the tip samples had been counted previously during August and the only one I could really get a reasonable decay curve from was the hottest sample collected on 19 January 1961, and this decayed with an apparent half-life of about 1½ years.

HOLLAND: I have a question I would like to ask Mr. FERBER on the fallout models. This is with regard to the other variables in the model. In particular, is there good agreement on the vertical distribution of radioactivity and distribution of radioactivity with particle size; you seem to have focused on the other variables? I would assume that all the models make essentially the same assumptions on these two.

FERBER: I would not say that we focused on other variables, but rather on the differences in the final dose predictions which result from the different assumptions as to the vertical distribution of radioactivity, the particle size distribution and so on.

HOLLAND: It is not a question of one model having provision for the shear and the other not. Actually, these differences are in the assumptions with regard to the vertical distribution and particle size distribution.

FERBER: Yes, the basic differences among the models lie in the assumed total amount of radioactivity, its distribution within the nuclear cloud and its distribution with respect to particle size. In this paper we have compared the final results rather than the individual assumptions. We also tried to indicate the magnitude of the effect of the meteorological parameters (wind speed and shear) on the fallout predictions.

Russel A. MARTIN (Hq. U. S. Air Force (AFCIN-3K2), Washington): I noticed you used primarily the 10 MT yield for these comparisons.

FERBER: We used 1 MT and 10 MT.

MARTIN: Did you use anything in the intermediate range? If so, did you see any wide discrepancies?

FERBER: We have looked at predictions ranging from 1 KT up to 20 MT. While the relationship between the various model predictions changes with weapon yield, the examples we have shown are typical. The predictions are not in good agreement at any yield.

ATMOSPHERIC INFLUENCES ON FALLOUT DEPOSITION

PART 2. ATMOSPHERIC MOTIONS

b. Global Fallout

Session Chairman: Joshua Z. Holland, Chief
Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission

AN INTERPRETATION OF GLOBAL FALLOUT

L. Machta, R. J. List and K. Telegadas
U. S. Weather Bureau*
Washington, D. C.

Introduction

This paper will cover the following subjects: An inventory of Sr^{90} prior to the U.S.S.R. 1961 nuclear tests; the goegraphical and temporal distribution of stratospheric fallout during the period 1959 to 1961 when very little of the fission product radioactivity was of tropospheric origin and finally a prediction of the Sr^{90} fallout from the U.S.S.R. 1961 tests.

The Inventory of Sr^{90} Prior to September 1961

There are two parts to the determination of the Sr^{90} inventory. First, the residual atmospheric content must be measured by filtration of the air at many places and altitudes in the atmosphere and second, the deposited Sr^{90} must be obtained by one of several techniques. We shall deal first with the atmospheric content.

In May 1960 the High Altitude Sampling Program of the Defense Atomic Support Agency was succeeded by a semi-annual program of monitoring in time and altitude at 4 latitudes. These are shown in Fig. 1 which displays the Sr^{90} concentrations in the atmosphere in November 1960. The data on this figure emphasize the broad governmental contributions largely instigated and supported by the Fallout Studies Branch of the U. S. Atomic Energy Commission (AEC) in measuring atmospheric radioactivity. Ground level observations are obtained on a monthly composite of daily filter collections made by the weather services along the 80th meridian (West) and analyzed by the U. S. Naval Research Laboratory.

* Research conducted under the auspices of the Fallout Studies Branch, Division of Biology and Medicine, U. S. Atomic Energy Commission.

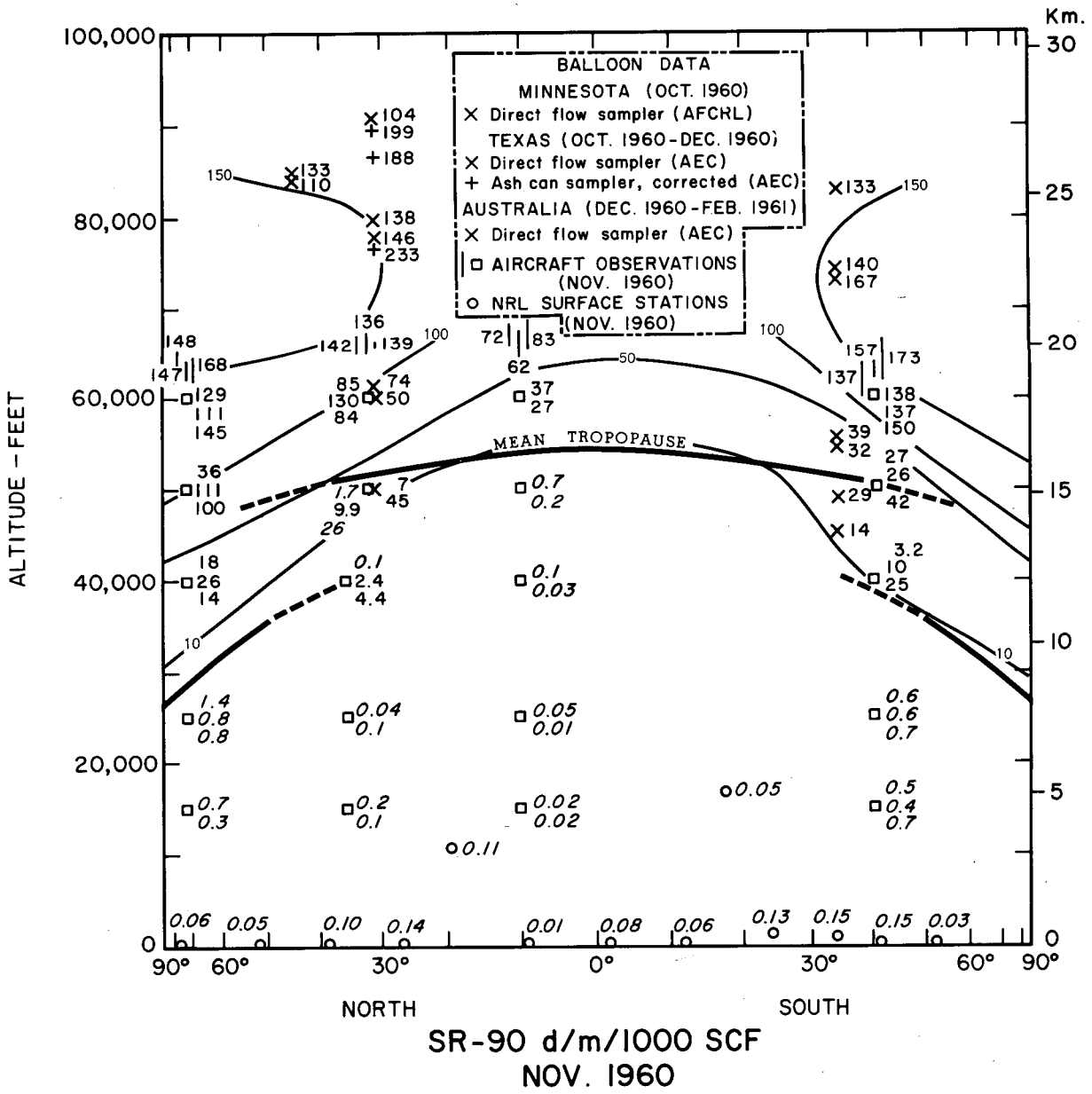

Figure 1. Global Atmospheric Distribution of Sr^{90} in November 1960.

B-57 aircraft were employed up to about 50,000 feet and U-2 aircraft above this altitude. The AEC balloon program results in Texas, Minneapolis, and Australia reach to 90,000 ft.

The heavy line (dashed in the temperate zones due to its transitory nature) represents the mean position of the tropopause, the imaginary boundary between the troposphere below and the stratosphere above. Note that the troposphere is deeper over the equatorial region and shallower over the polar areas. The tropopause in the temperate and polar regions also rises in the spring and falls in autumn and varies in height with the weather situation. The lines shown in the figure are grossly idealized; actual, real daily pictures vary greatly from the average and frequently show more than a single gap in each hemisphere. In some instances, as may be inferred from the diagram, a station may have two (or more) tropopause surfaces above it.

The figure clearly shows that the main concentrations of Sr^{90} lie in the stratosphere. There is a change of concentrations from values of 1 or less in the troposphere to almost 200 in the stratosphere. There is also a large gradient in concentration just above the average tropopause with values at 40,000 and 50,000 ft. intermediate between true tropospheric values and true stratospheric values.

The picture also reveals smaller equatorial stratospheric concentrations than farther to the north or south. The concentrations in the Southern Hemisphere stratosphere appear to be about the same as in the Northern Hemisphere. Finally, there is only a minor suggestion of any decrease in concentration with altitude at the balloon stations.

Fig. 2 shows a similar Sr^{90} air concentration pattern about 6 months later, the last survey which was made before the resumption of atmospheric tests by the U.S.S.R. in September 1961. Most of the features described above for the November 1960 picture apply to the May 1961 pattern as well. A few differences can be noted. First, concentrations in the Northern Hemisphere have decreased, including perhaps even those at 65,000 ft. over equatorial stations. But up to 70,000 ft. in the Southern Hemisphere, the stratospheric concentrations have generally remained as high as before or have even increased. This leads to the surprising pattern of greater Southern than Northern Hemisphere stratospheric concentrations despite the fact that effectively all of the stratospheric Sr^{90} injections have taken place in the Northern Hemisphere. Second, the north polar tropospheric concentrations (as well as the entire Northern Hemisphere tropospheric content) have risen since November. This result is in accord with our previous experience as will be reported below. It is unfortunate that the changes in stratospheric concentrations cannot be examined for possible seasonal effects since it is presumed that the Northern Hemisphere stratosphere is thoroughly contamined by the 1961 U.S.S.R tests.

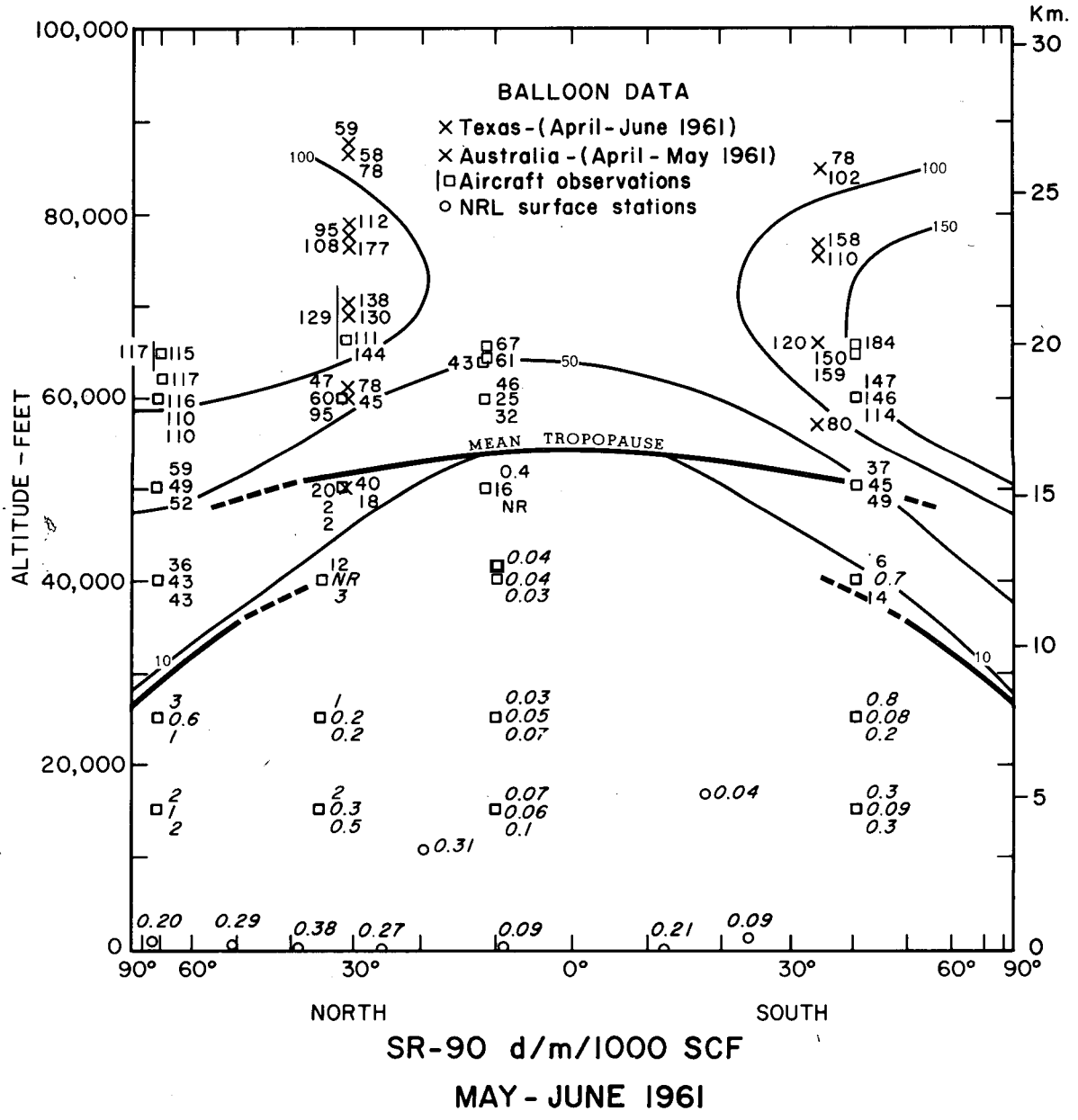

Figure 2. Global Atmospheric Distribution of Sr^{90} in May - June 1961.

It is a relatively simple mechanical procedure to compute the atmospheric burden of Sr^{90} from Figs. 1 and 2 or similar charts. The results are listed in Table 1 for 3 dates. The stratospheric inventory is subdivided by altitude and hemisphere. The Sr^{90} content above 100,000 ft. is not based on direct observation. Rather, it is assumed that the Rh^{102} (to be discussed later) is a tracer for the 0.4 Mc of Sr^{90} which are presumed to have been inserted into the mesosphere by the U. S. Johnston Island rocket tests (during Operation HARDTACK I, 1958). From the amount of Rh^{102} below 100,000 ft. one may deduce the amount of Sr^{90} still left above the sampling altitude. The inventory calculation between 70,000 and 100,000 ft. one may deduce the amount of Sr^{90} still left above the sampling altitude. The inventory calculation between 70,000 and 100,000 ft. is seriously hampered by the lack of equatorial observations. It has been assumed that the Sr^{90} concentration continues to increase above 70,000 ft. as suggested by the ascendant between the tropopause and 70,000 ft. Finally, it can be noted that the surprising excess of Sr^{90} at the Southern Hemisphere aircraft sampling station over its Northern Hemisphere counterpart actually reverses the ratio of Southern to Northern Hemisphere content in this layer between May-June 1960 and May 1961.

The deposited Sr^{90} can be obtained from soil and rainfall analyses. This is illustrated in Fig. 3 in which the Sr^{90} deposition is plotted against latitude, on an equal area scale. The dashed curve is derived from Alexander et al. (1) to mid-1959. This is supplemented by data from the AEC pot and ion exchange collection of rainfall. The shaded area portrays the increment from mid-1959 to May 1961. This figure can be integrated and the results appear in the line of Table I entitled "Deposited."

The sum of the Sr^{90} in the "Total Atmosphere" and that "Deposited" on the earth's surface represents the accountable Sr^{90}. As of May 1961, this equals about 5.7 Mc. The totals have not changed significantly since May 1960 which is consistent with the small ($2\frac{1}{2}\%$) yearly decay.

This inventory calculation may be compared with one obtained independently. The AEC has announced that approximately 9.2 Mc of Sr^{90} (based on the assumption that 10 MT of fission energy equals 1 Mc of Sr^{90}) have been created by nuclear tests before November 1958. It is estimated, but with considerable uncertainty, that about 3.0 Mc fell locally on or about test sites. The remaining6.2 Mc have been corrected for decay to the appropriate dates and listed along the last line of Table I for comparison with the observed inventories. The agreement is very good. This is fortuitious, perhaps, since there is evidence that the oceanic fallout may exceed that over land by as much as a factor of three (2). Further, in both the atmospheric and deposited inventory, the number and distribution of observations leaves much to be desired.

Geographical Distribution of Stratospheric Fallout

It is evident in Fig. 3 that the total Sr^{90} fallout is non-uniformly

TABLE 1

Sr^{90} Inventories (Mc)
(Decayed to Indicated Date)

	May 1960		November 1960		May 1961	
	Northern Hemisphere	Southern Hemisphere	Northern Hemisphere	Southern Hemisphere	Northern Hemisphere	Southern Hemisphere
Above 100,000ft (est).	0.14	0.14	0.13	0.13	0.12	0.12
70-100,000 ft.	0.23	0.23	0.20	0.20	0.14	0.14
Tropopause-70,000	0.25	0.19	0.24	0.24	0.19	0.26
Stratosphere	0.62	0.56	0.57	0.57	0.45	0.52
Troposphere	0.03		0.02		0.03	
Total Atmosphere	1.21		1.16		1.00	
Deposited	4.50		4.61		4.73	
Total	5.7		5.8		5.7	
World-wide debris*	5.5		5.4		5.3	

* Based on 9.2 Mc less 3.0 Mc of local fallout.

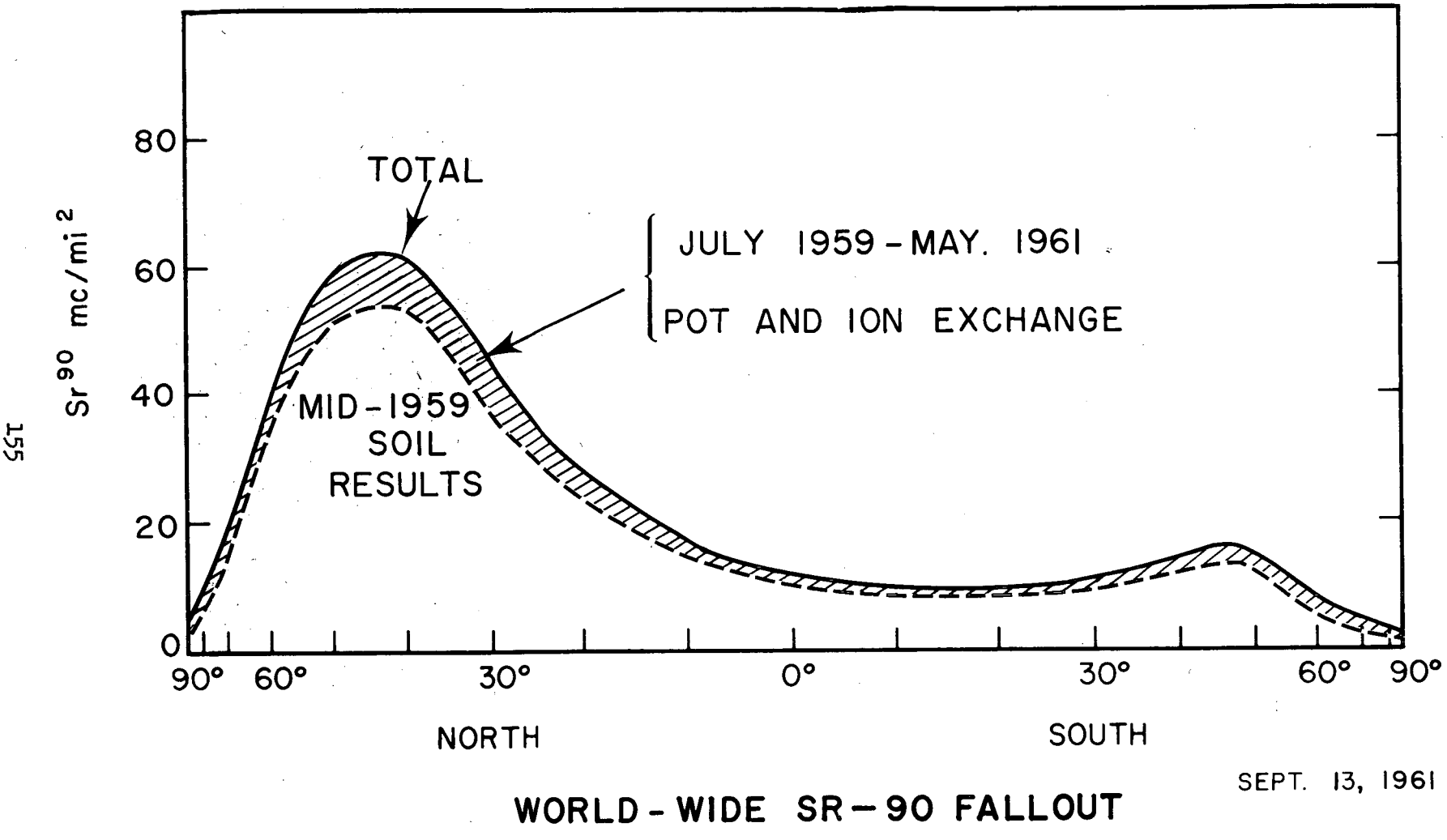

Figure 3. Global Distribution of Deposited Sr^{90} up to May 1961.

distributed over the earth's surface. Actually, the graph fails to reveal a second type of irregularity. This results from precipitation variations. The smoothness of the profile in Fig. 3 is a consequence of averaging around circles of latitude and additional smoothing.

The most obvious feature is the peak in the North Temperate Zone and the secondary maximum in the South Temperate Zone. These maxima are not the consequence of greatest precipitation in these regions, although the decrease towards the poles may reflect the lesser precipitation in the Arctic and Antarctic regions. Thus, for example, there is heavy rainfall in the equatorial belt which has less observed fallout than that to the north or south.

Fig. 3 reflects the total Sr^{90} fallout from all sources to date. Part of this fallout is of tropospheric origin (never having entered the stratosphere), part from U. S. and United Kingdom tests near the equator and part from U.S.S.R. tests closer to the north pole. In the next two figures, it will be shown that the main features evident from the total fallout appear in the fallout pattern from injections in either the equatorial or polar stratosphere. This is of considerable significance both to the health physicist and the meteorologist.

Fig. 4 shows a north-south profile of Sr^{90} deposition in periods of a most exclusively stratospheric fallout. The early 1959 total fallout, the solid line, is seen, in the Northern Hemisphere, to be mostly composed of October 1958 U.S.S.R. debris, the dashed line. Both curves show a very marked North Temperate Zone peak. It is further noted that a curve of the residual deposition, that is, the fallout from sources other than October 1958, would also peak in the North Temperate Zone. The dotted line shows the total fallout in the first half of 1960 when it is estimated that only a very small portion of the fallout could be attributed to the October 1958 U.S.S.R series. Again a peak in the North Temperate Zone is apparent.

It can be inferred from Fig. 4 that the distribution of fallout from stratospheric sources is not uniform. The marked peaking in the North Temperate Zone in early 1959 is mainly but not entirely due to the U.S.S.R. October 1958 test series. But even when the October 1959 fallout contribution is relatively small, as in early 1960, there is still a maximum in the North Temperate Zone.

In Fig. 5, one finds the geographical distribution of W^{185}. Approximately 100 Mc, corrected for decay to 1 June 1958, were injected into the lower equatorial stratosphere by the HARDTACK test series. In all likelihood most of this tracer material was inserted below 80,000 ft. However, a fraction of the W^{185} was left behind in the trosphere during the conduct of the tests. Consequently, the pattern of W^{185} fallout from the HARDTACK tests due to the stratospheric component is obtained by selecting the fallout in the AEC pots and ion exchange columns subsequent to January 1959. By this date it is believed that essentially all of the tropospheric component of the W^{185} would have been washed out of the atmosphere.

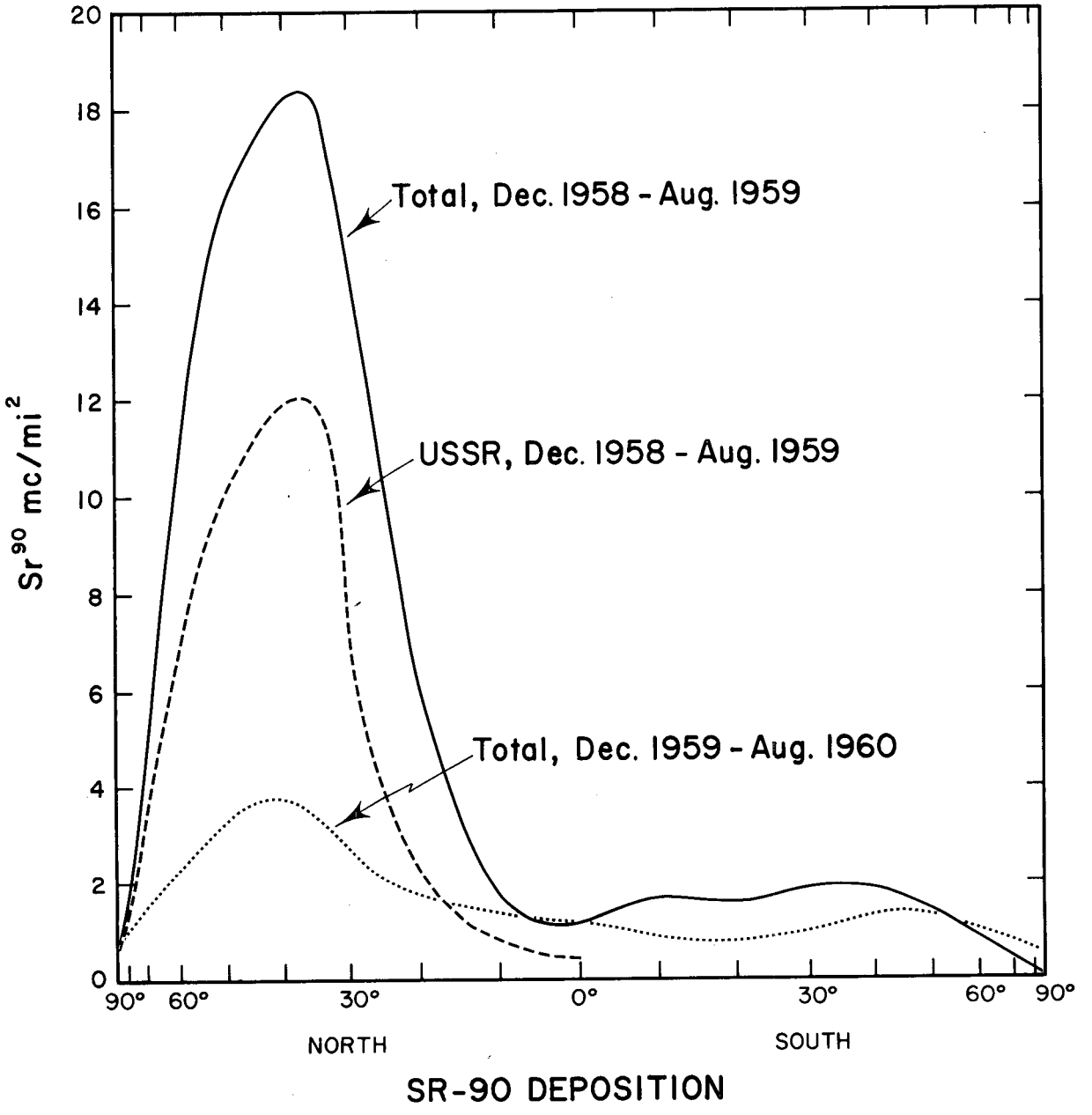

Figure 4. Global Distribution of Deposited Sr^{90} During the "Spring" of 1959 and 1960 Showing Also the Fallout from the U. S. S. R. October 1958 Tests in the "Spring" of 1959.

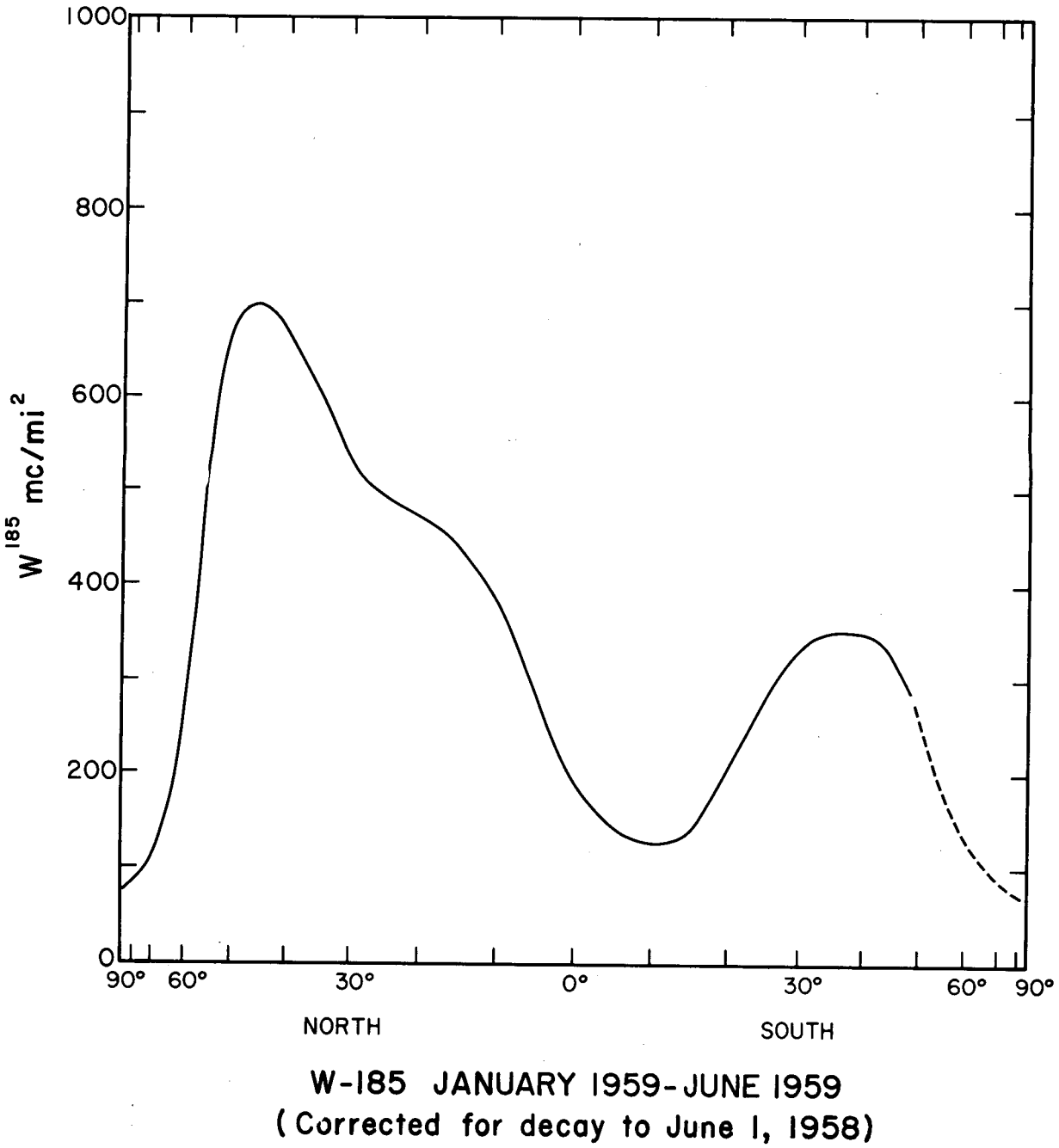

Figure 5. Global Distribution of W^{185} During the First Half of 1959.

Fig. 5 shows once again the marked peak in the North Temperate Zone. In this case, the peak in the South Temperate Zone is also evident, although there is practically no information to show that a decrease occurs southward to the South Pole from the South Temperate Zone peak. Thus, despite the fact that the source of the W^{185} originated at 11°N, the stratospheric fallout shows the same geographical distribution found in Figs. 3 and 4 from all sources and from the Arctic source.

In summary, it is concluded that irrespective of the source of the stratospheric radioactive aerosols, the distribution of fallout and ground level air concentration is characterized by a peak in the Temperate Zones. Certain reservations should be noted. First, there have been numerous instances of ground level air concentrations remaining as high or increasing toward the Arctic from the North Temperate Zone. Second, there is no unequivocal tracer for radioactive fallout originating at about the 100,000-ft. level. Such injections occurred in the IVY (1952) or CASTLE (1954) tests. It has been estimated from pre-1956 fallout data that this, too, peaks in the Temperate Zones.

Seasonal Distribution of Stratospheric Fallout

Fig. 6 is a plot of the Sr^{90} fallout and air concentrations for each hemisphere based, in the upper part of the figure, on the AEC pot and ion exchange collections and, in the lower part of the figure, on the Naval Research Laboratory's 80th meridian network air concentration measurements. It is apparent that the Northern Hemisphere curves reveal peaks in the spring and troughs in the fall. The Southern Hemisphere curves, on the other hand, do not show the same systematic seasonal trends. However, in the Southern Hemisphere spring of 1960 and autumn of 1961, one does find a reasonable suggestion of a parallel to the Northern Hemisphere history. It should also be noted that it is only during this interval that the air concentrations in both hemispheres are about equal; during all earlier periods the concentration of Sr^{90} was appreciably higher in the Northern Hemisphere than in the Southern Hemisphere. This difference will have a bearing on an explanation of the absence of a seasonal variation in the Southern Hemisphere to be given below.

The Sr^{90} reported in Figure 6 includes fallout from all nuclear tests. A breakdown into indentifiable geographical sources of stratospheric fallout is given in the next table and two figures.

Table 2 lists the number of Mc of Sr^{90} deposited in 3-monthly intervals subsequent to the October 1958 U.S.S.R. tests in the indicated regions of the globe as deduced from the AEC pot and ion exchange observations. The lower line in each set of data is the fallout attributed to the U.S.S.R. October 1958 tests determined from an assumed activity Sr^{89}/Sr^{90} ratio of 170 at time of formation, a date of formation of

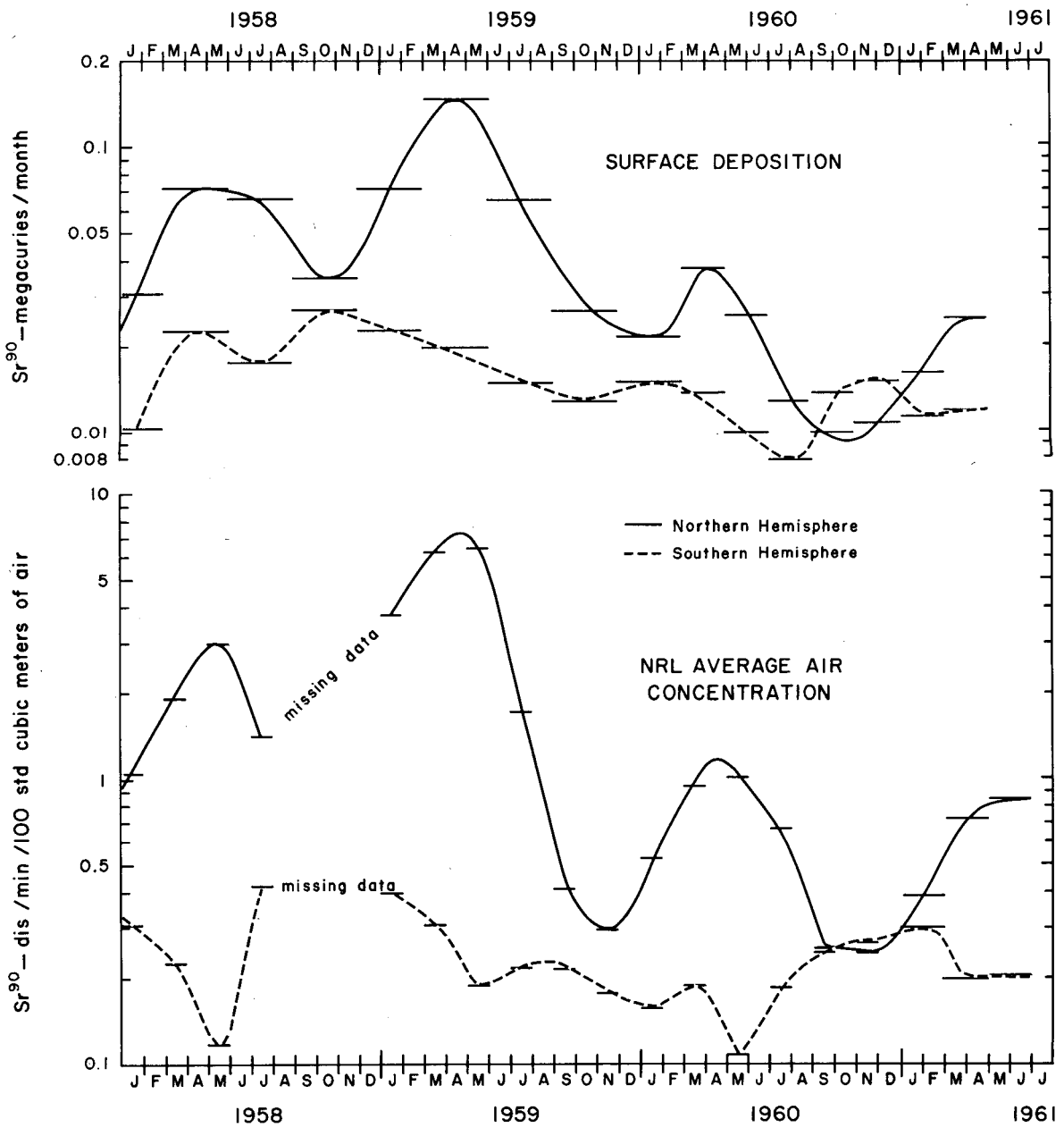

Figure 6. Seasonal Trends of W^{185} in the Northern and Southern Hemisphere from a Stratospheric Equatorial Source.

TABLE 2

World-wide Sr^{90} Deposition from November 1958 to December 1959
and U.S.S.R. Contribution (Based on Sr^{89}/Sr^{90} Data)

		Sr^{90} Deposition (Mc)				
		(N) 90-30	(N) 30-0	(N) 90-0	(S) 90-0	Total
Dec. 1958	Total Deposition	0.15	0.06	0.21	0.07	0.28
Feb. 1959	U.S.S.R.	0.11	0.03	0.14	0.01	0.15
Mar. -	Total Deposition	0.34	0.12	0.46	0.05	0.51
May 1959	U.S.S.R.	0.27	0.07	0.34	0.01	0.35
June -	Total Deposition	0.16	0.04	0.20	0.03	0.23
August 1959	U.S.S.R.	0.10	0.02	0.12	0.01	0.13
Sept. -	Total Deposition	0.06	0.03	0.09	0.03	0.12
Nov. 1959	U.S.S.R.	0.02	0.01	0.03	0.01	0.04
Dec. 1958	Total Deposition	0.71	0.25	0.96	0.18	1.14
Nov. 1959	U.S.S.R.	0.50	0.13	0.63	0.04	0.67

October 20, 1958, and the assumption that all of the Sr^{89} is derived from these U.S.S.R. tests. Errors in these assumptions can alter the absolute amounts of Sr^{90} attributed to the U.S.S.R. test series but is unlikely to modify the seasonal trend.

The table shows that period March-May 1959 contained the largest amount of fallout both in terms of total fallout and that ascribed to the U.S.S.R. test series in the Northern Hemisphere. The decrease during the summer and fall of 1959 is marked. Most of the seasonal trends appears in temperate and polar bands (north of 30°N). There is no seasonal trend in the Southern Hemisphere. An analysis by Gustafson (3) suggests a similar seasonal trend in the spring of 1960 for the U.S.S.R. 1958 contribution with the air concentration at Argonne National Laboratory being about two orders of magnitude less in the spring of 1960 than in the spring of 1959.

Fig. 7 displays the time history of air concentrations of W^{185} derived from the Naval Research Laboratory's 80th meridian network in both hemispheres. Recall that the sources of the W^{185} is the lower equatorial stratosphere in the summer of 1958. A peak value in the spring of 1959 is followed by a minimum in the autumn of 1959 and a second peak in the spring of 1960 in the Northern Hemisphere. The Southern Hemisphere simply shows a generally downward trend with a suggested bump in the late winter of 1959. Again, it should be noted that the Northern Hemisphere concentration is significantly greater than the Southern Hemisphere concentration, as was the case during the greater part of the period included in the Fig. 6.

Fig. 8 shows the time history of Rh^{102} at the only 2 stations at which measurements of this nuclide are available. This figure is made available through the courtesy of Dr. P. F. Gustafson of Argonne National Laboratory. However, the 1959 results are in doubt and must be considered subject to correction upon reanalysis by Dr. Gustafson. It should be pointed out that while an estimated 3.0 Mc of Rh^{102} were produced by the high altitude Orange test at Johnston Island on August 12, 1958, there were also about 0.3 Mc created by the HARDTACK tests in the summer of 1958 which left their debris in the lower equatorial stratosphere. For this reason, there can be ambiguity concerning the source of any Rh^{102} observed in small amounts in 1959. It was not until September 1959 (4) that the Rh^{102} could be clearly identified as having originated at high altitudes, both from the amounts involved and from its stratospheric distribution. The results in 1960 and 1961 appear to be beyond question at the moment.

Fig. 8 reveals a seasonal trend in the Southern Hemisphere with a spring maximum and autumn minimum. (In spite of the remarks of the above paragraph, it may still be that the Santiago peak in November 1959 may be real and due to the high altitude source. Even if due to the lower altitude equatorial source, the presence of a peak in November is relevant.) The measurements near Argonne National Laboratory show the expected seasonal variation as found in other types of observations. Thus, almost for the first time, there appears to be good agreement between the seasonal trends in both hemispheres.

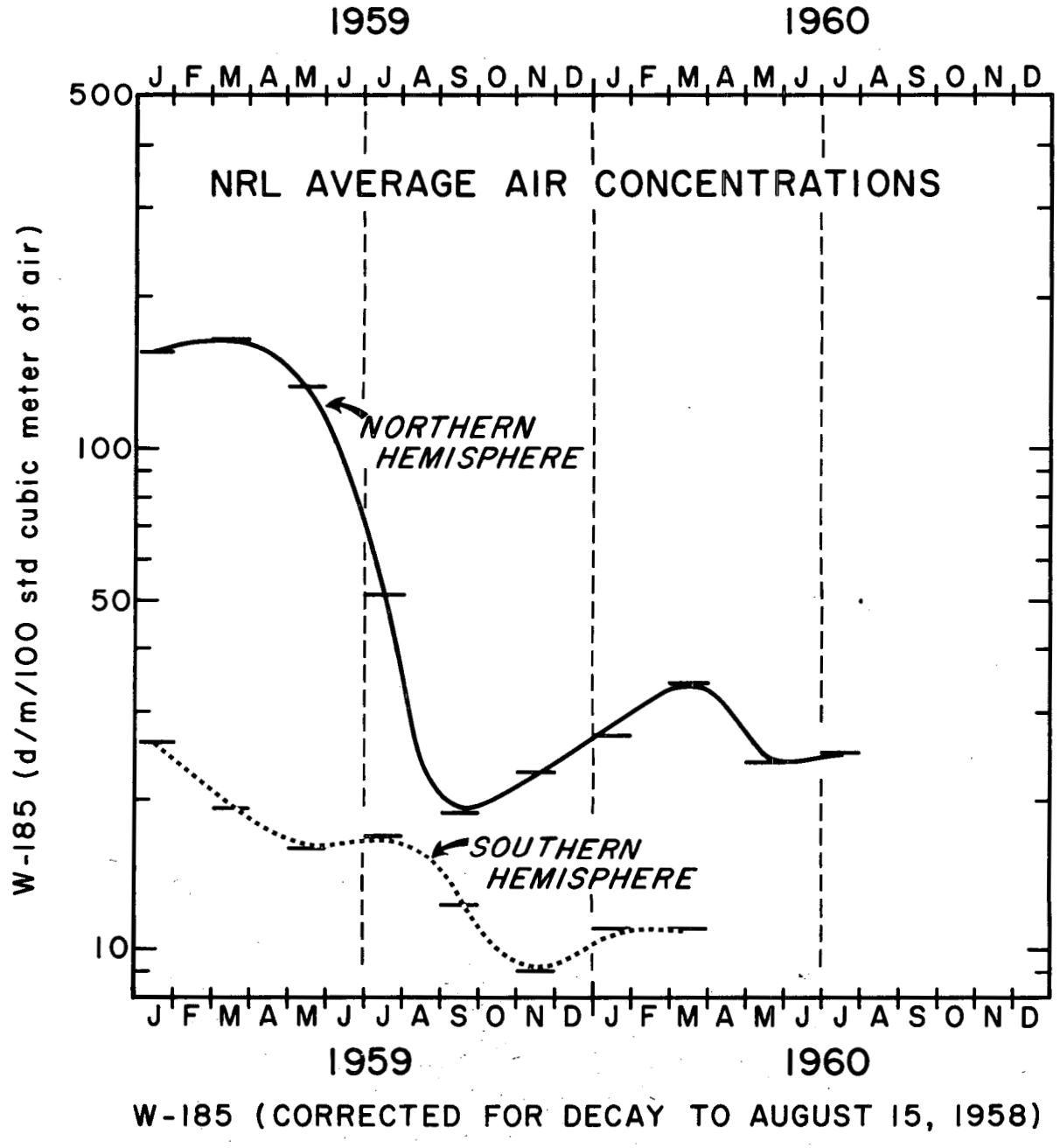

Figure 7. Seasonal Trend of W^{185} in the Northern and Southern Hemisphere from a Stratospheric Equatorial Source.

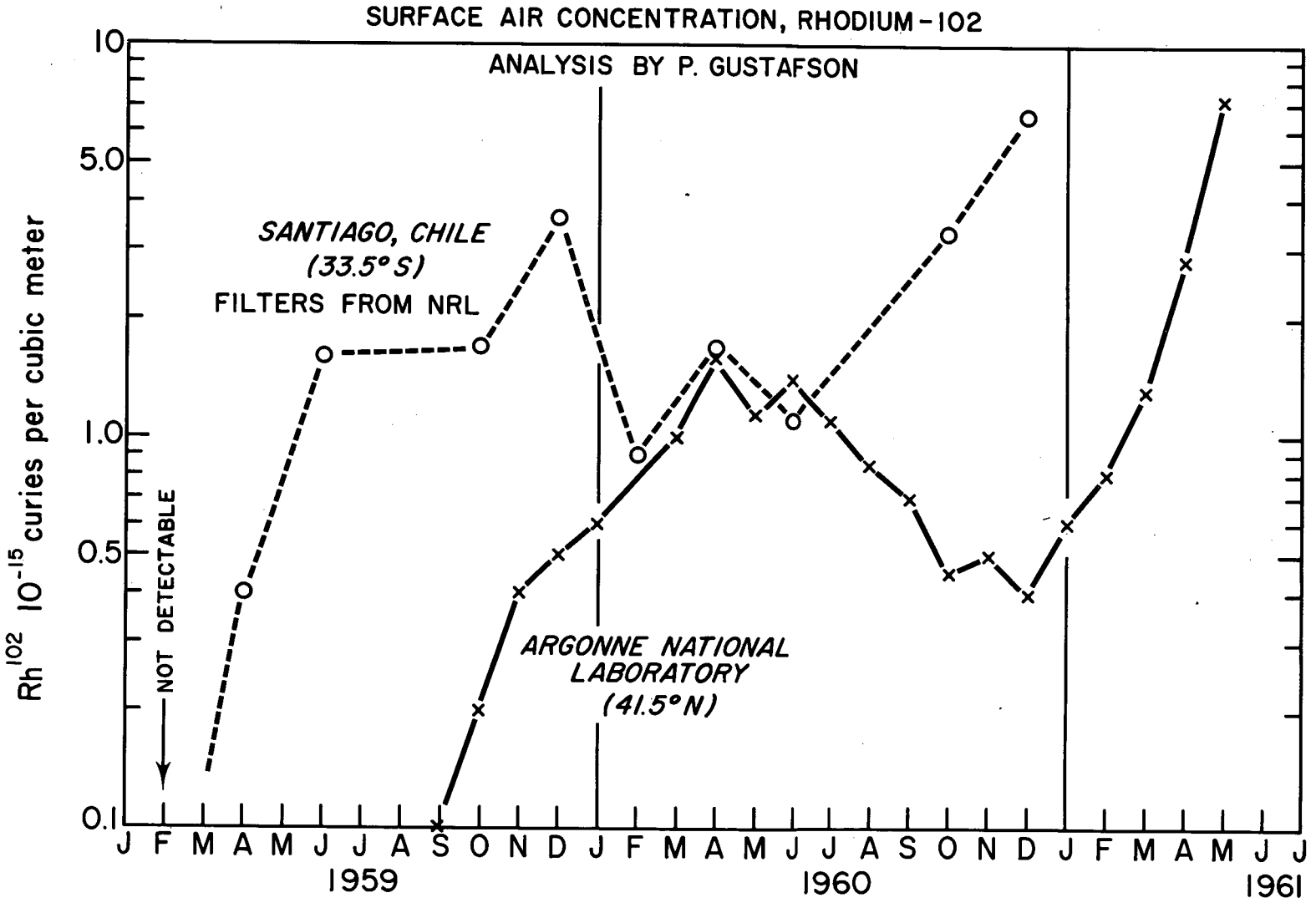

Figure 8. Seasonal Distribution of Rh^{102} at a Northern and a Southern Hemisphere Station.

It can be noted that the levels of Rh^{102} concentration are equal in both hemispheres. An equality between hemispheres was previously noted in 1960-1961 in the Sr^{90} fallout and air concentration measurements when a suggestion of a seasonal trend was likewise found.

It is here proposed that removal from the stratosphere in both hemispheres takes place seasonally with a maximum in late winter or possibly early spring. However, there is also cross-equatorial mixing occurring in the upper troposphere from the hemisphere of higher concentrations (the Northern) to the hemisphere of lower concentration (the Southern). This cross-equatorial tropospheric exchange forms a second source of tracer material for the Southern Hemisphere such that the "expected" seasonal variation in ground level air or fallout in the Southern Hemisphere may be confused. It is only when a tracer of approximately equal concentration exists in each hemisphere that the seasonal trend in the surface concentrations can be detected. Thus, had not the U.S.S.R. 1961 tests again produced large amounts of long-lived fission products for entry into the Northern Hemisphere troposphere one might have expected to have found a regular Southern Hemisphere seasonal variation in years subsequent to 1961. Fig. 2 showed that the stratospheric content of the Southern Hemisphere was about as great in the Southern as Northern Hemisphere just before September 1961.

It should be recognized that the above simple explanation for the absence of a seasonal variation of fallout in the Southern Hemisphere in most data is a matter of speculation. There are a number of real differences in in the structure and behavior of the atmosphere in the two hemispheres which could also explain the differences in fallout. For example, the unexpected excess of Sr^{90} in the Southern Hemisphere lower stratosphere over that in the Northern Hemisphere lower stratosphere in 1961 undoubtedly is indicative of such differences. The possible difference in radioactive fallout behavior between hemispheres due to meteorological processes must be studied more extensively before the explanation given in the previous paragraph can be accepted.

Predictions of Future Fallout From the U.S.S.R. Autumn 1961 Tests

The AEC has announced that the total fission yield of the U.S.S.R. test series in the autumn of 1961 equalled 25 MT of equivalent energy. This is about twice the fission yield of the U.S.S.R. October 1958 test series. On this basis, one might double the fallout attributed to the October U.S.S.R. 1958 tests as has been done in Fig. 9. The peak fallout of Sr^{90} in Fig. 4, for the time period ("spring" 1959) which is argued to have included most of the U.S.S.R. October 1958 fallout, appears to be significantly less than half the average concentration at about the 40-45°N peak in

Fig. 9. The reason for this differences lies in methods of analysis of the data. In Fig. 4, the observed pot and ion exchange results from land-based stations were processed objectively (including taking into account rainfall over the ocean) with the resulting fallout of about 0.7 Mc of U.S.S.R. October 1958 debris instead of almost 1.25 Mc. In Fig. 9 the fallout over land stations from which soil, pot and ion exchange collections were available strictly scaled by a factor of 2. But over the oceans where rainfall and fallout observations are poor, the Sr^{90} fallout was increased in accordance with recent work by Bowen and Sugihara (2) such that the total deposited Sr^{90} in Fig. 9 was forced to equal approximately 2.5 Mc.

As noted in the section on the seasonal distribution of fallout, almost all of the fallout from tests conducted by the U.S.S.R. in the Arctic is deposited during the first spring following the explosions. This would again be expected of the U.S.S.R 1961 tests if the altitude of injection were the same. Fig. 10 shows a plot of the tops of nuclear clouds versus the total yield of the explosion (fission plus fusion). Thus, clouds from tests near the ground whose yield is several MT might be confined to the atmosphere below 80,000 to 90,000 ft. During the 1961 test series, 2 tests were conducted which had yields in excess of several MT; a 25 and a 55-60 MT explosion. If the mean (heavy) curve in Fig. 10 is extrapolated to these yields, it is clear that there should be a sizeable amount of the debris between 100,000 and 150,000 ft. It is believed that this is considerably greater than the height of stabilization of the debris from the 1958 tests. This greater height would possibly have the effect of delaying the time of fallout. Thus, contrary to the rapid fallout in 1959 from the October 1958 tests, it could take several years for the same fraction of stratospheric fission products to be deposited. However, it is still expected that well over 50% of the U.S.S.R. stratospheric debris will be deposited in the first spring (1962). This is based in part on the AEC statement that "Of special interest is the small fission yield of the 50-60 megaton test conducted on October 30" which indicates that the fission products from this test may not have contributed significantly to the 25 MT of fission yield.

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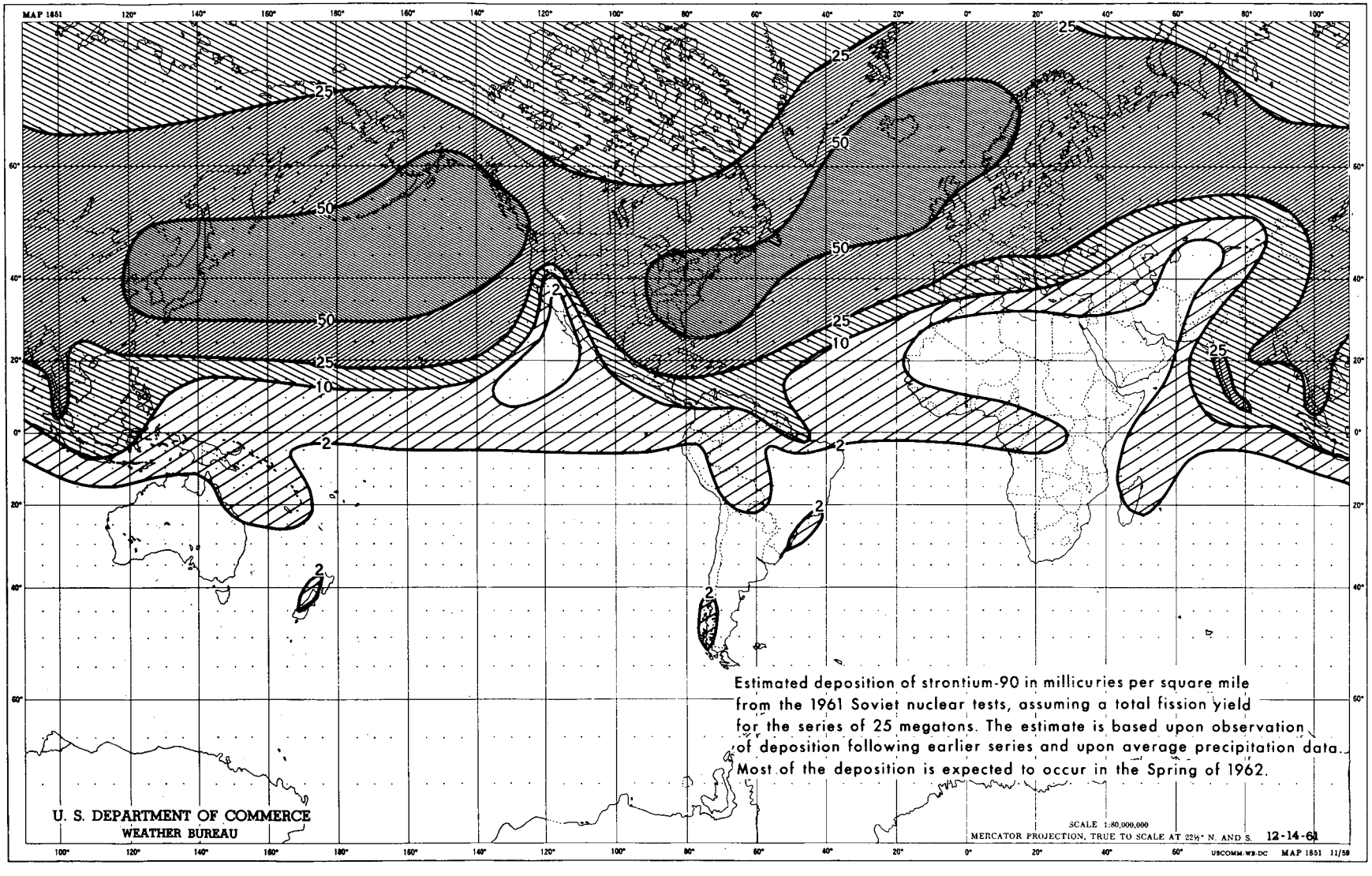
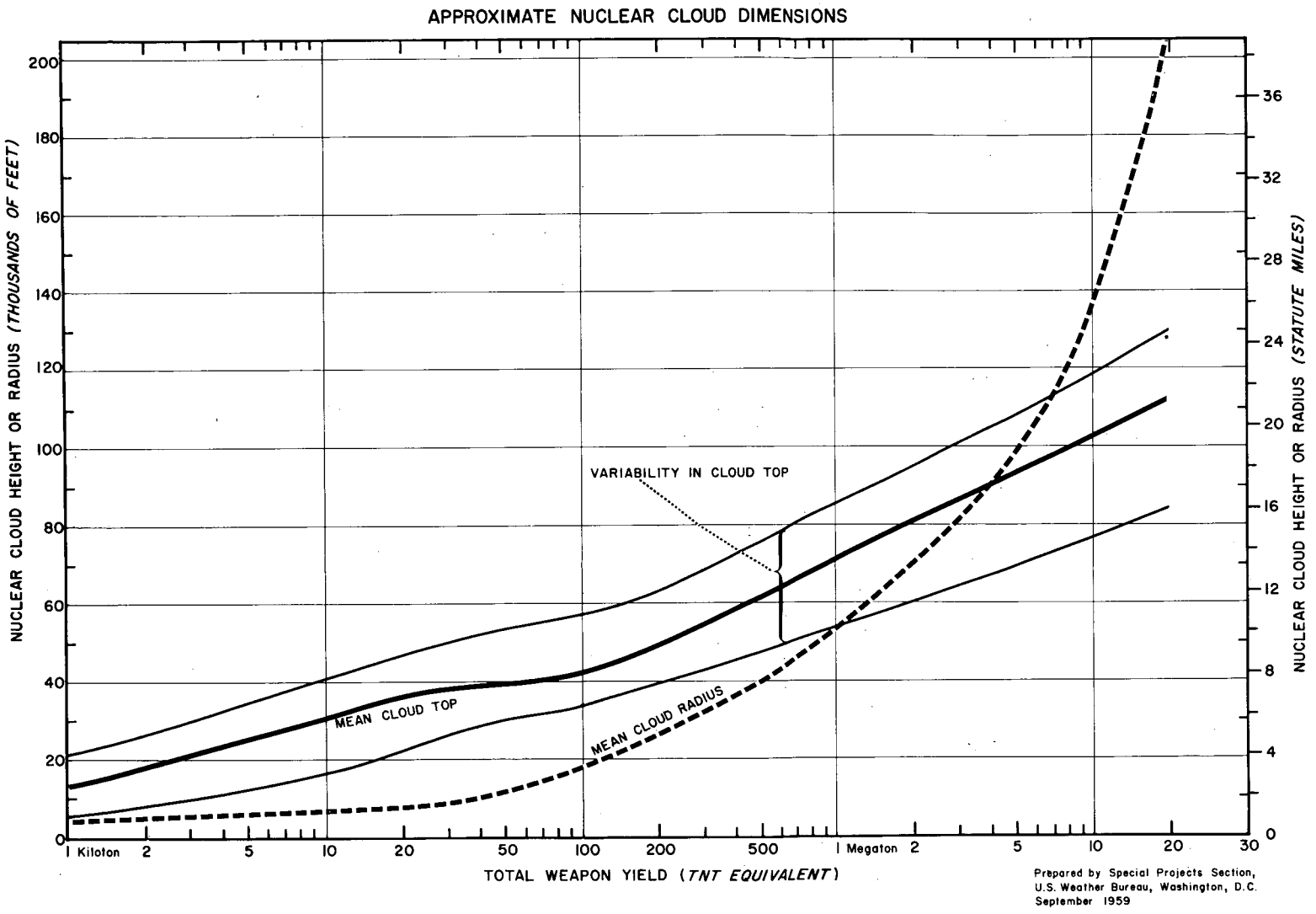


Figure 9. A Prediction of Fallout from the Autumn 1959 U. S. S. R. Test Series.



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PRELIMINARY RESULTS ON THE SIZE AND VERTICAL DISTRIBUTIONS OF RESIDUAL NUCLEAR DEBRIS IN THE STRATOSPHERE

Peter J. Drevinsky and Edward A. Martell
Air Force Cambridge Research Laboratories
Cambridge, Massachusetts

Introduction

The study of the distribution of stratospheric contamination and the mechanisms of transport involved have been hampered by the paucity of good stratospheric data. On the one hand, there has been no adequate evaluation of physical properties of radioactive aerosols in the stratosphere without which the role of sedimentation cannot be elucidated. In addition, data on the stratospheric radioactivity distribution above U-2 aircraft ceilings of about 20 km are both sparse and of uncertain quality. The need for data over the high equatorial tropopause where U-2 aircraft can at best make only shallow penetrations is of special importance in connection with the evaluation of the role of organized meridional circulation in debris transport.

In an attempt to satisfy these requirements, we have initiated an investigation of the physical properties of stratospheric aerosols as a function of altitude using a balloon-borne impactor-filter sampling system. In addition, measurements of fission products and natural radioactivities were carried out at Hyderabad, India, as part of a Joint United States-India Balloon Program, during the period February through April 1961. Preliminary results of these particle size studies and of the India balloon profile radioactivity measurements are presented below.

Techniques

The particle size studies were carried out with the large volume impactor unit, which was originally designed and calibrated in cooperation with General Mills, Inc., under Contract No. AF 19(604)-4943. This collection device, having a capacity of 1000 ambient cu. ft. of air per sample, is essentially a scaled-up version of the impactor air sampler used by Chagnon and Junge (1, 2) in their particle size studies of natural aerosols. In the large unit, four separate barrels are manifolded to the one pumping system and individually programmed during the ascent to provide a four-point vertical profile on a single balloon flight. The altitude ranges are 30 to 50, 50 to 70, 70 to 90, and 90 to 100 thousand feet. Each barrel has a two-stage impactor, and a third-stage polystyrene microfiber filter as a back-up collector for the very small particles. The impactor jets were individually designed and calibrated with homogeneous aerosols at the mid-point of the particular altitude range, so that the first stages have a 50% collection efficiency of 0.15 μ radius, and the second stages 0.02 μ radius, assuming a reasonable value for the particle density of 2 g/cm^3. These cutoffs were kept fairly uniform regardless of altitude by increasing the jet dimensions with altitude to compensate for the decrease in air density, according to theoretical considerations. In addition, a similar impactor-filter device, constructed for the U. S. Atomic Energy Commission by General Mills, Inc., was employed for some single-level collections in the lower stratosphere. This instrument, with only one barrel, has jets calibrated at 0.18 μ and 0.008 μ radius, but the volume capacity of 25 cu. ft/min. and other design features are the same.

The larger balloon-borne air filter samples collected in the Hyderabad, India flights were obtained using the Air Force Cambridge Research Laboratories Sub-Micron Aerosol Collector (SMAC). This air filter unit employs 10 ft.^{2} of polystyrene microfiber filter material in a cylindrical configuration similar to that of the original Ash Can sampler. The design and operational characteristics of this sampler have been described in a recent General Mills, Inc., report (3). The polystyrene filter material used was produced by Stanford Research Institute (SRI) under Contract No. AF 19(604)-2644. The method of fiber production and other characteristics of the filter material are reported elsewhere (4).

The filter fibers of the SRI paper range from about 0.1 to 1.0 μ diameter. Tests on this paper indicate essentially 100% efficiency for collection of sub-micron aerosols over a wide range of particle sizes and flow rates (4, 5).

Preliminary treatment of the 10 ft.^{2} polystyrene filter samples included the pelletizing of 1 ft.^{2} fractions in a press, addition of pellets and appropriate carriers to a 500 ml distillation flask, and destructive distillation of the polystyrene material. The small residues containing the natural and artifical radioactivities of interest were wet-ashed with a

a sulfuric and nitric acid mixture and analyzed by standard radiochemical separation procedures.

Impactor-Filter Results

The total beta activity results for impactor-filter samples collected over Minneapolis, Minnesota (45°N, 93°W), and Hyderabad, India (17°N, 79°E), are summarized in Table 1. The beta activity measurements for each set of data were carried out in a reproducible manner, with a geometry factor of about 3.1, and are reported in counts per minute (cpm) per thousand cubic feet, ambient. The percentage of activity in each size fraction and in each altitude interval is presented in Fig. 1 for two profiles over Minneapolis, Minnesota, and in Fig. 2 for two profiles over Hyderabad, India. The samples were collected about two years or more after the last nuclear test and sample volumes were quite small. The activity of only a few samples was adequate for individual nuclide measurements. Results of Ce^{144} and Sr^{90} activity measurements for these samples are summarized in Table 2.

Figures 1 and 2 show that most of the activity in the lower stratosphere is on the second-stage of the impactor, indicating the predominance of activity in the 0.02 to 0.15 μ radius particle size range. Perhaps the most striking feature is the marked altitude trend for particles of the smallest fractions. Some 60 to 80% of the activity at altidues of 90,000 to 100,000 ft. is distributed in particles of sizes less than 0.02 μ radius. The anomaly indicated by one low altitude sample over Minneapolis and another over Hyderabad, showing more activity on the first stage, is unexplained. It may be due either to a small shift in the effective cutoff of the impactor first stage or to an actual shift in radioactivity-aerosol size distribution. Additional data on the size distribution of radioactive particles between 15 and 21 km altitude over Minneapolis are shown in Fig. 3. Again, most of the activity is on the second stage, with the exception of one flight for which the first stage impactor cutoff was adjusted downward to 0.05 μ radius. For the latter, the concentration of activity on the first stage indicates that most of the activity must be in the narrow range of approximately 0.05 to 0.15 μ radius, the lower end of the natural sulfate aerosol size distribution at this level (6).

The limited individual nuclide data, Table 2, were obtained to check on the possible physical and chemical fractionation of individual radioisotopes with particle size. The accuracy of the analyses of the Ce^{144} and Sr^{90} activities was limited by the small sample size. Within experimental uncertainties, there is no indication of a variation of fission product composition with altitude or with particle size. However, the data are inadequate to assess the possibility of moderate variations of this nature.

TABLE 1

Radioactivity of AFCRL impactor-filter collections [a]

Collection date	Counting date	Altitude (10^3 ft.)	Total Beta cpm/10^3 ft.^{3}, Ambient Stage 1[b]	Stage 2[b]	Filter
Minneapolis, Minnesota:					
7 Jun. 60	18 Aug. 60	30-50	6.40 ± 0.12	0.105 ± 0.025	0.586 ± 0.084
		50-70	1.38 ± 0.05	36.0 ± 0.2	4.47 ± 0.12
		70-90	0.288 ± 0.058	13.7 ± 0.2	7.54 ± 0.14
		90-100	0.096 ± 0.063	2.78 ± 0.16	3.74 ± 0.24
11 Oct. 60	20 Nov. 60	30-50	0.655 ± 0.067	3.10 ± 0.09	≤0.45
		50-70	3.84 ± 0.10	31.6 ± 0.4	1.86 ± 0.16
		70-90	1.25 ± 0.09	17.8 ± 0.2	6.03 ± 0.23
		90-100	0.823 ± 0.089	1.60 ± 0.12	4.07 ± 0.25
15 Feb. 61	17 Mar. 61	67	5.96 ± 0.08	11.5 ± 0.1	2.72 ± 0.04
16 Jun. 61	6 Jul. 61	65	38.6 ± 0.3	5.78 ± 0.09	0.485 ± 0.035
Hyderabad, India:					
13 Apr. 61	12 May 61	50-70	0.951 ± 0.079	4.94 ± 0.12	≤0.65
		70-90	2.56 ± 0.08	≤0.20	1.99 ± 0.11
		90-92.7	≤0.16	≤0.16	≤0.72
20 Apr. 61	25 May 61	50-70	1.45 ± 0.15	7.98 ± 0.24	≤1.6
		70-90	1.50 ± 0.15	10.4 ± 0.2	5.72 ± 0.35
		90-97.9	≤0.71	2.45 ± 0.20	11.5 ± 0.5

a. The sampling system consists of a 2-stage impactor with backup filter, designed in cooperation with General Mills, Inc., under Contract No. AF 19(604)-4943. This system is an enlarged version of that used by Chagnon and Junge (1,2).

b. Impactor cutoffs were 0.15 and 0.02 μ radius for vertical profiles, 0.18 and 0.0075 μ radius for 15 February 1961 flight, and 0.05 μ and 0.025 μ radius for 16 June 1961 flight.

TABLE 2

Ce^{144} and Sr^{90} activity of impactor-filter samples

Flight Date	Altitude (10^3ft	Fraction	Concentration on Sampling Date (dpm/10^3ft^3, ambient) Ce^{144}	Sr^{90}	Ce^{144}/Sr^{90}	Total beta[a] (cpm)	Ce^{144}[b] (dpm/10^3ft^3, ambient)	$\frac{Ce^{144}\text{ (dpm)}}{\text{Total beta (cpm)}}$
7 Jun. 60	50-70	Stage 2	44.5 ± 1.3	5.70 ± 0.71	7.81 ± 0.99	36.0 ± 0.2	37.9 ± 1.1	1.05 ± 0.03
		Filter	4.72 ± 0.38	---	---	4.47 ± 0.12	3.97 ± 0.32	0.887 ± 0.075
	70-90	Stage 2	22.7 ± 0.5	2.07 ± 0.66	11.0 ± 3.5	13.7 ± 0.2	19.3 ± 0.5	1.41 ± 0.04
		Filter	8.10 ± 0.70	---	---	7.54 ± 0.14	6.76 ± 0.59	0.897 ± 0.094
	90-100	Filter	7.05 ± 0.77	---	---	3.74 ± 0.24	5.85 ± 0.60	1.56 ± 0.19
11 Oct. 60	50-70	Stage 2	36.2 ± 1.4	5.98 ± 0.66	6.05 ± 0.70	31.6 ± 0.4	32.6 ± 1.3	1.03 ± 0.04
		Filter	2.73 ± 0.50	---	---	1.86 ± 0.16	2.50 ± 0.46	1.34 ± 0.27
	70-90	Stage 2	21.7 ± 1.6	2.60 ± 0.52	8.34 ± 1.77	17.8 ± 0.2	19.2 ± 1.5	1.08 ± 0.08
		Filter	5.69 ± 0.83	---	---	6.03 ± 0.23	5.15 ± 0.75	0.854 ± 0.127
	90-100	Filter	4.91 ± 0.72	---	---	4.07 ± 0.25	4.39 ± 0.64	1.08 ± 0.16
15 Feb. 61	67	Stage 1	3.00 ± 0.12	0.924 ± 0.252	3.25 ± 0.89	5.96 ± 0.08	3.93 ± 0.16	0.660 ± 0.028
		Stage 2	8.04 ± 0.24	2.12 ± 0.10	3.79 ± 0.21	11.5 ± 0.1	10.5 ± 0.3	0.913 ± 0.027
		Filter	2.68 ± 0.10	0.608 ± 0.082	4.40 ± 0.62	2.72 ± 0.04	3.50 ± 0.14	1.29 ± 0.06

a. Total beta cpm value on counting date (see Table 1).

b. Ce^{144} value corrected for decay to total beta counting date.

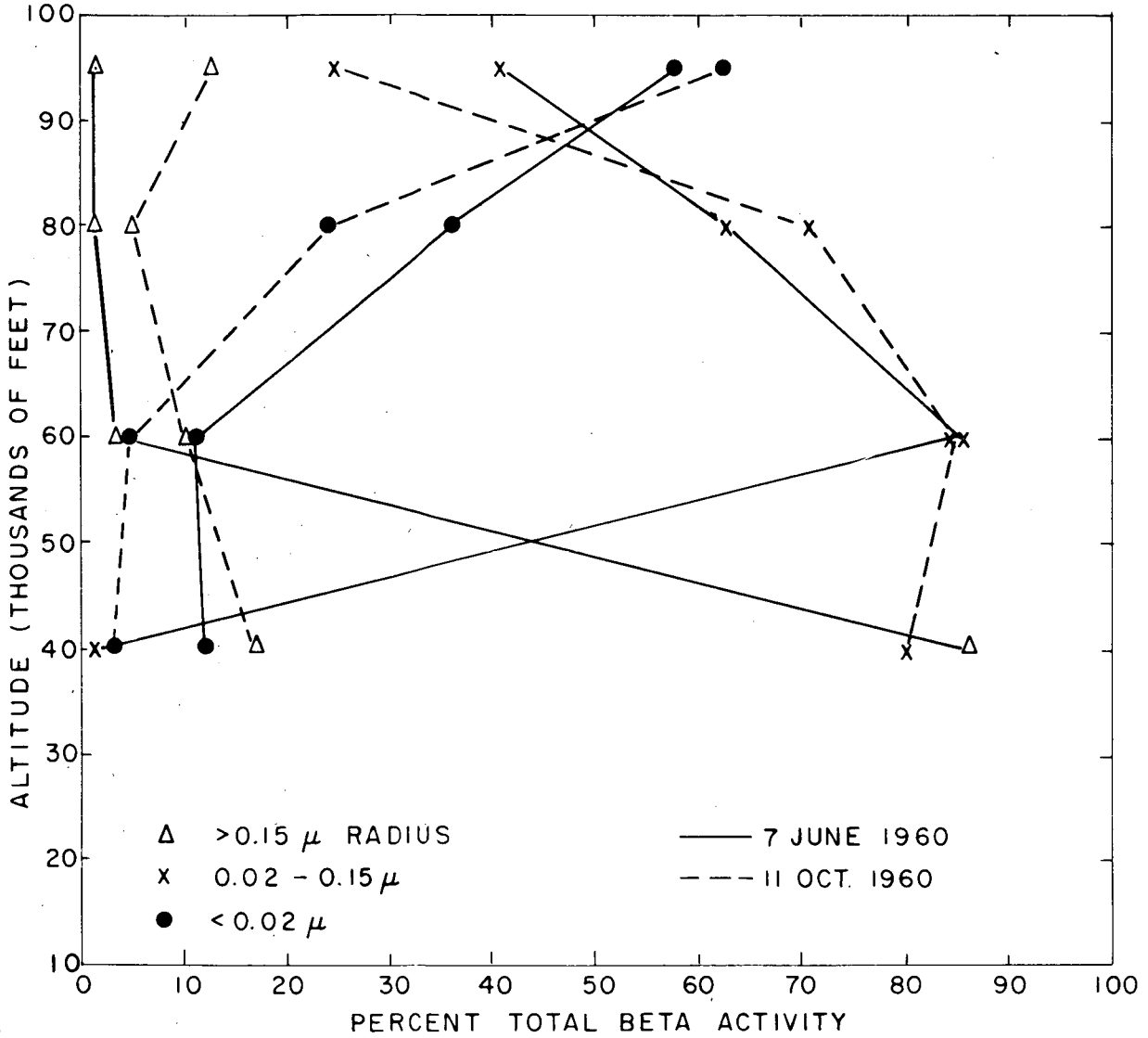

Figure 1. Fission Product Activity Fraction vs. Particle Size and Altitude Interval, Minneapolis, Minnesota (45°N, 93°W).

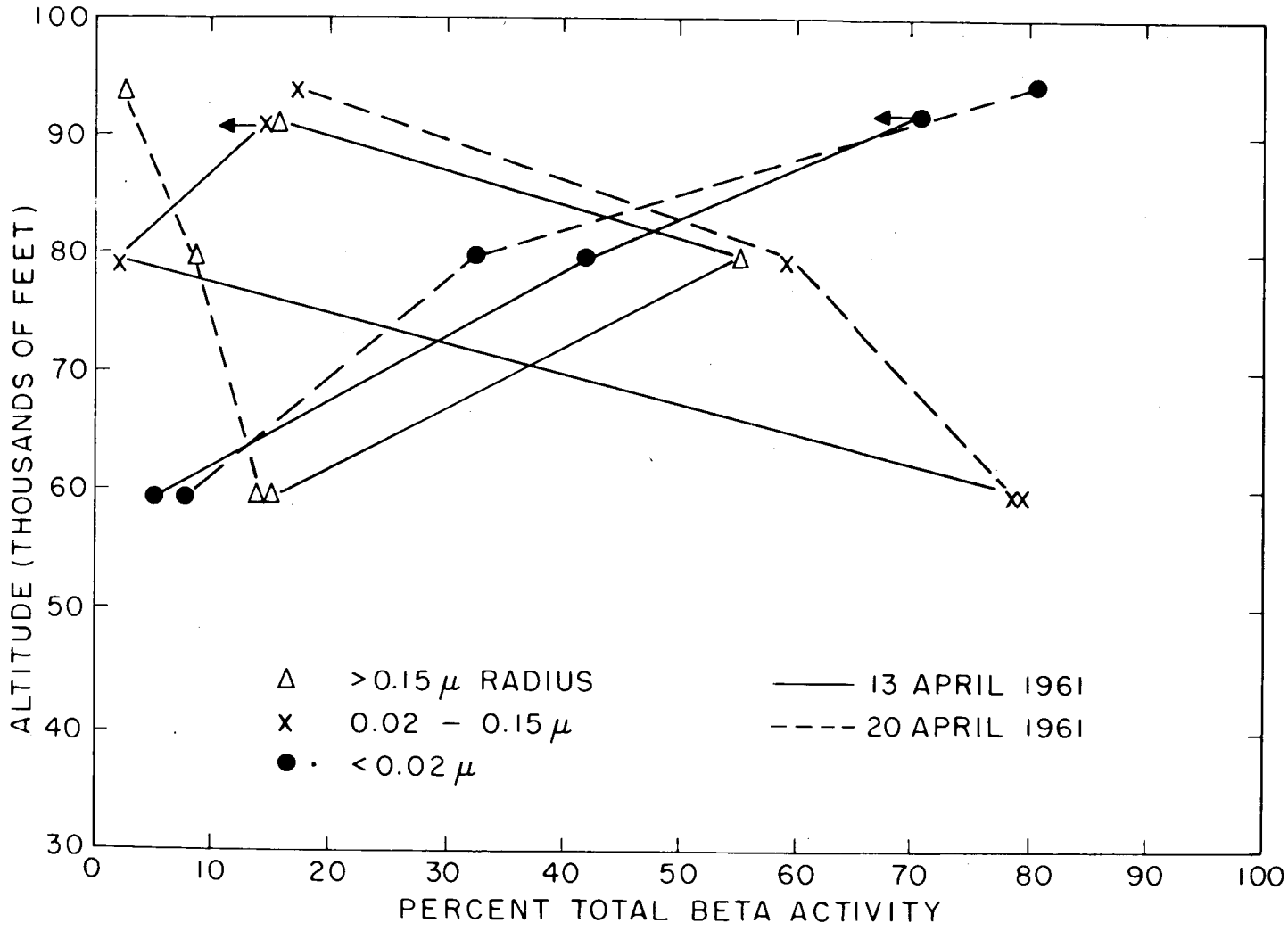

Figure 2. Fission Product Activity Fraction vs. Particle Size and Altitude Interval, Hyderabad, India (17°N, 79°E).

Stratospheric Radioactivity Over Hyderabad, India

Flight information and radioactivity data for the submicron aerosol collector samples obtained over Hyderabad, India, are presented in Tables 3 and 4. Volume data, Table 3, provided by General Mills, Inc. (GMI), were based on both PR-2 flowmeter and blower rpm data. Unfortunately, the results are inconsistent and unreliable. Flowmeter failures are indicated at least in the case of flights 2522, 2525, and 2535. For the other flights, the flowmeter also leads to an underestimation of the actual volume sampled. On the other hand, blower rpm data at best provide only an upper limit of the air sample volume. In this case, any obstruction to a free flow of air (twisted duct, obstructed entrance, or other such malfunctions) would result in a reduced air-flow which would not be reflected by the rpm data.

The measurements of Ce^{144}, Sr^{90}, Rh^{102}, and Pb^{210} presented in Table 4 were carried out with care. The ratios Ce^{144}/Sr^{90} are approximately the values to be expected for fission products produced during 1958, with small variations outside of counting statistics. The variations may be due to somewhat incomplete mixing of sources of different age or of different fission-product production ratio, or both. The Rh^{102}/Sr^{90} ratios show a fairly systematic trend with altitude, with an increase by a factor of about 4 over the altitude range between the lower stratosphere and 97,000 ft. The activity concentrations given in Table 4, based on rpm volume data, show unexpected large variations in concentration at a given altitude. Because variations of this sort are inconsistent with the expected distribution of fission products in the equatorial stratosphere 2.5 years after production, they are attributed mainly to volume uncertainties.

To establish a vertical distribution of fission products and Rh^{102} over Hyderabad, the total beta activity profile for the 20 April 1961 impactor-filter flight affords the only reasonable possibility. The impactor-filter unit employs a constant volume displacement pump which allows accurate assessment of volumes on the basis of pumping speed, pump temperature, and pressure-altitude data. The reproducibility of this method is shown by the two profiles over Minneapolis, Minnesota, Table 5 and Fig. 4, as well as by the sulfate aerosol profile data of Junge, Chagnon, and Manson (6). The 20 April 1961 activity profile over Hyderabad, India, is also shown in Table 5 and Fig. 4. Unfortunately, this was the only impactor-filter flight over India for which reliable volume data exists.

The activity profiles, Fig. 4, are necessarily distorted because samples were taken over extended altitude intervals. The multiple impactor-filter units were neither designed nor intended for vertical profile measurements. The profiles in Fig. 4 were drawn with the assumption that all of the activity for the lowest altitude interval was collected

TABLE 3

GMI volume data for SMAC flights over Hyderabad, India.

Flight No.	Sampling altitude (ft.)	Sampling time (min.)	Sampling rate ($ft.^3$/min.)	Ambient volume ($ft.^3$)	Volume S.T.P. ($ft.^3$)	Method
2521	76,500	93	542	50,380	1,980	Flowmeter
			543	50,500	1,980	rpm
2522	97,000	153	---	--	--	Flowmeter
			317	49,100	693	rpm
2525[a]	89,470	120	136	16,350	330	Flowmeter
			777	93,300	1,890	rpm
2534	68,530	79	354	27,900	1,650	Flowmeter
			618	48,800	2,890	rpm
2535[a]	77,360	139	27.9	3,880	147	Flowmeter
			530	73,600	2,790	rpm
2536	86,800	120	627	75,200	1,730	Flowmeter
			482	57,900	1,330	rpm
2537	97,300	101	480	48,600	730	Flowmeter
			372	37,600	560	rpm

a. High blower exhaust temperatures noted.

TABLE 4

Radioactivity of SMAC samples, Hyderabad, India, March-April 1961, based on rpm volume data

Flight No.	1961 Date	Altitude (ft)	Volume S.T.P.(a) ($ft.^3$)	Activity ($dpm/10^3 ft.^3$, S.T.P.) Ce^{144}(b)	Sr^{90}	Rh^{102}(c)	Pb^{210}
2521	13 March	76,500	1,983	440 ± 4	100 ± 8	71.4	≤0.6
2525	22 March	89,470	1,886	39.2 ± 1.3	9.17 ± 0.80	<17.5	≤0.35
2522	14 March	97,000	693	384 ± 4	59.7 ± 5.9	169	≤0.7
2534	5 April	68,530	2,890	103 ± 2	32.2 ± 1.7	≤14	--
2535	6 April	77,360	2,790	102 ± 2	16.9 ± 1.1	≤9.6	--
2536	7 April	86,800	1,330	553 ± 4	161 ± 6	~156	--
2537	8 April	97,300	560	362 ± 8	111 ± 5	~168	--

a. Volumes, based on rpm data, are in doubt and on the high side.

b. Ce^{144} data corrected to 26 March 1961.

c. Rh^{102} data corrected to 12 August 1958.

TABLE 5

Total beta activity vs. altitude for impactor-filter profiles

A. Minneapolis, Minnesota (45°N, 93°W)

(1) Flight 2468, 7 June 1960, tropopause height 39,600 ft.

Altitude (10^3 ft.)	Volume, ambient ($ft.^3$)	Volume S.T.P.[a] ($ft.^3$)	Total Beta (G x cpm/10^3 $ft.^3$ S.T.P.)
30-50	1132	262	96 ± 3
39.6-50	589	98	254 ± 10[a]
50-70	1314	119	1430 ± 10
70-90	1110	36	2050 ± 32
90-100	616	9.2	1370 ± 65

(2) Flight 2488, 11 October 1960, tropopause height 47,000 ft.

30-50	833	196	53 ± 4
47-50	125	17.6	590 ± 64[a]
50-70	743	70	1220 ± 20
70-90	624	22	2200 ± 30
90-100	523	7.2	1460 ± 70

B. Hyderabad, India (17°N, 79°E)

Flight 2542, 20 April 1961, tropopause height 53,000 ft.

50-70	374	35.4	334 ± 28
53-70	318	27.7	427 ± 37[a]
70-90	313	11.1	1540 ± 37
90-97.9	240	3.0	3550 ± 150

(a) Assumes activity collected above tropopause.

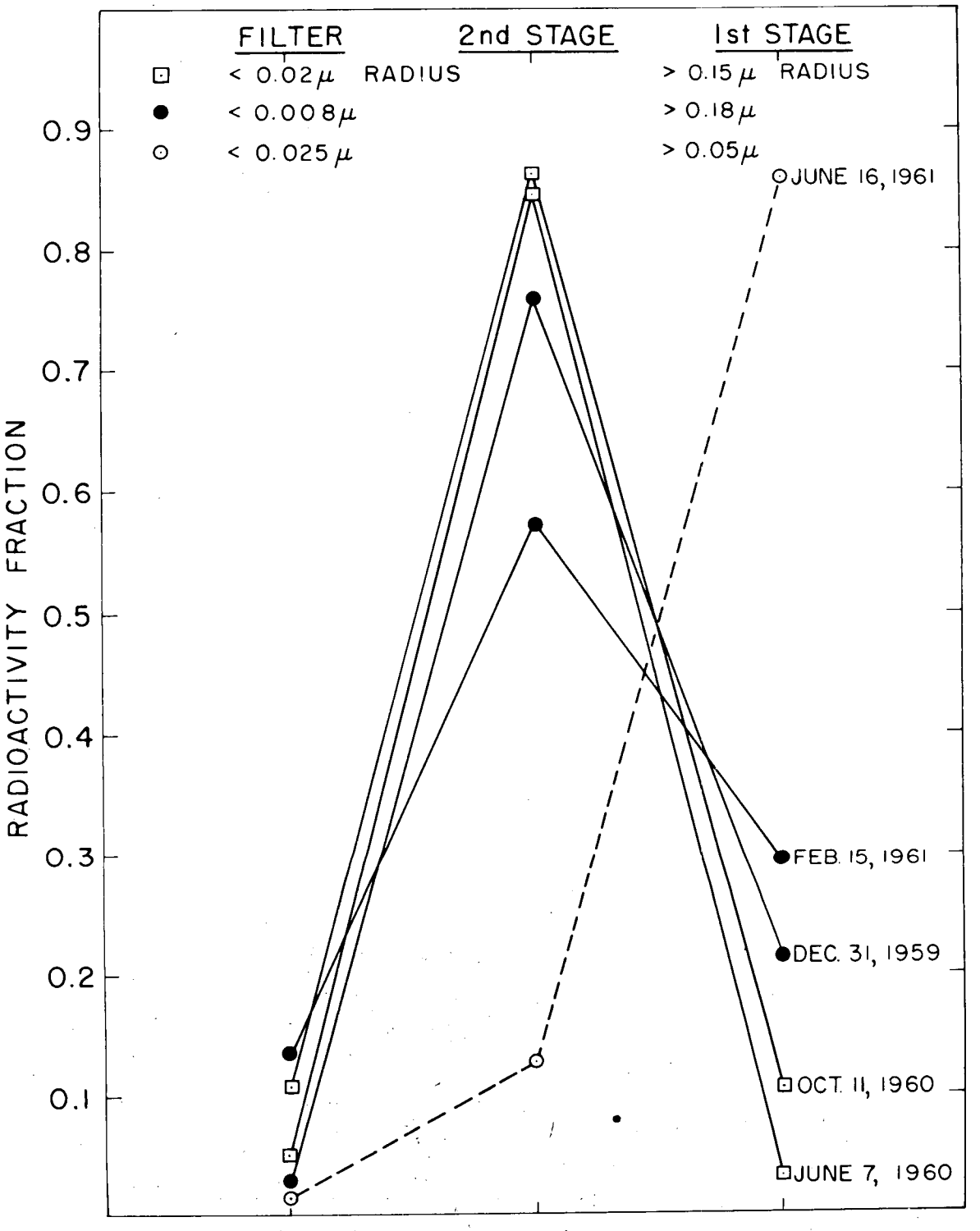

Figure 3. Radioactivity Distribution of Impactor-Filter Collections at 15-21 Kilometers over Minneapolis, Minnesota (45°N, 93°W), at Several Times and With Varied Cutoff.

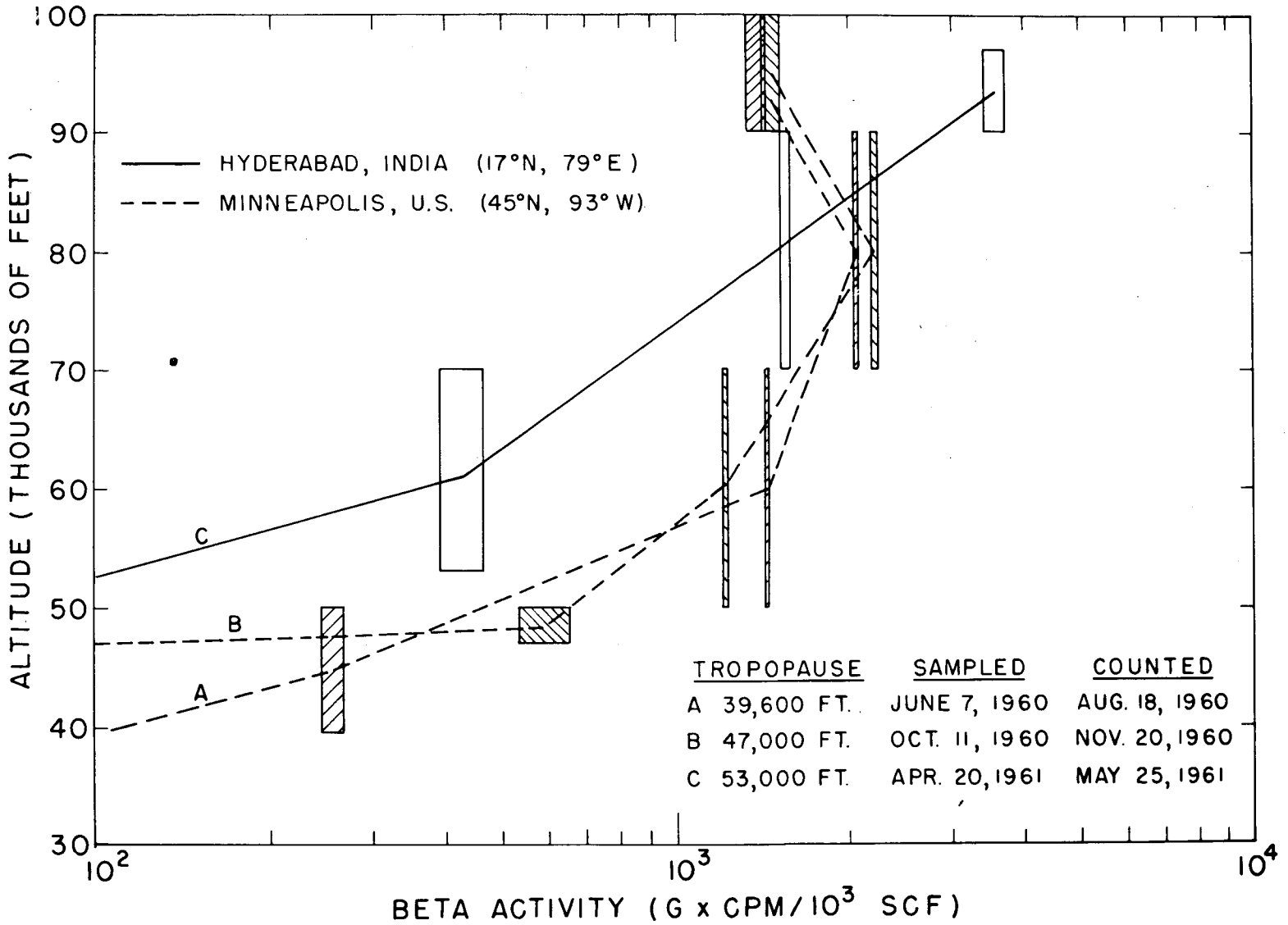

Figure 4. Fission Product Beta Activity vs. Altitude from Impactor-Filter Profile Flights over Minneapolis, Minnesota, and Hyderabad, India.

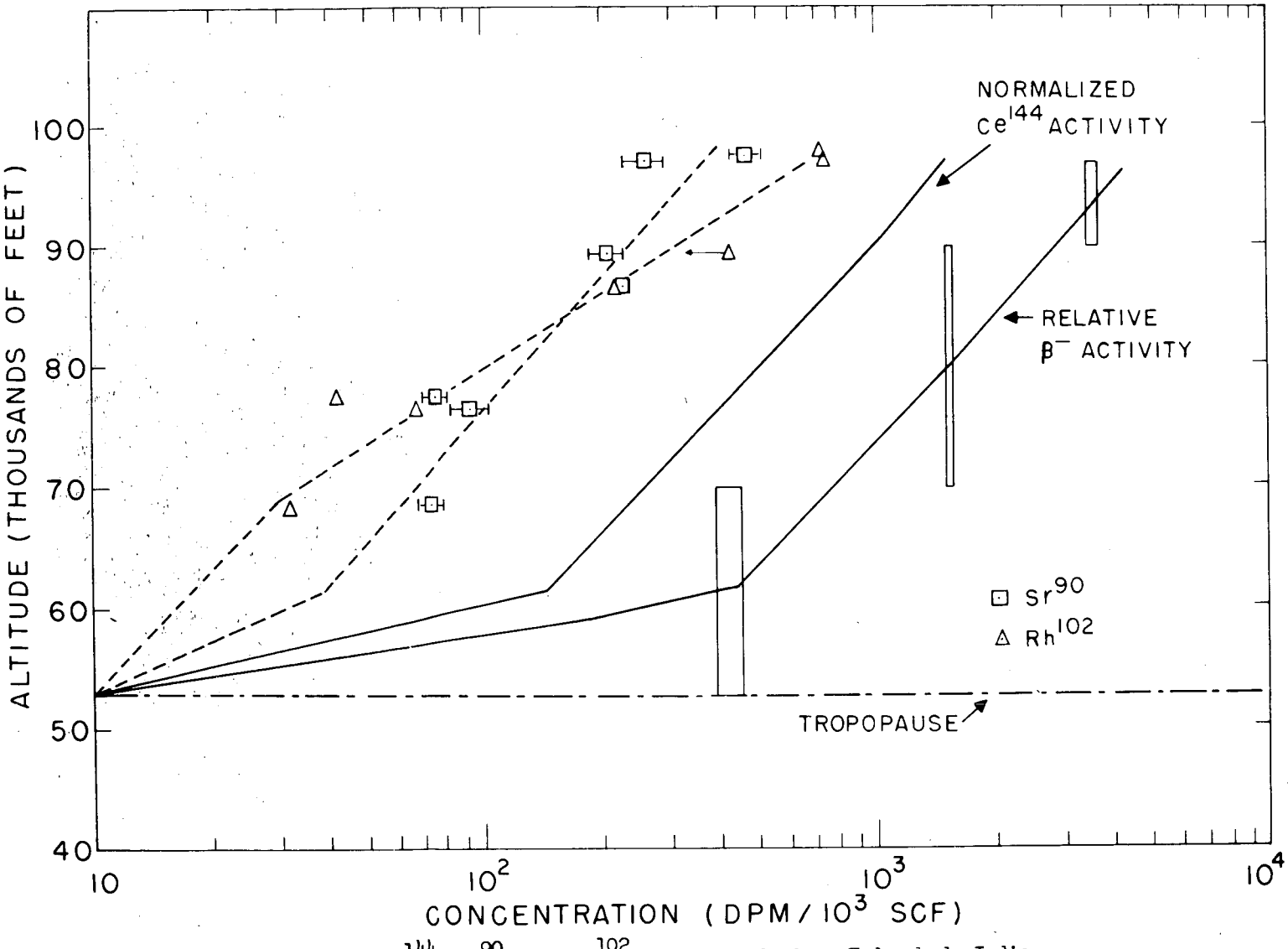

Figure 5. Ce^{144}, Sr^{90}, and Rh^{102} *vs.* Altitude Over Hyderabad, India (17°N, 79°E), Based on Impactor-Filter Profile, 20 April 1961.

above the tropopause. For the India profile, Fig. 4, it appeared to be more appropriate to connect points of mid-altitude rather than mean pressure altitude in view of the indicated exponential increase of activity vs. altitude.

The Ce^{144}, Sr^{90} and Rh^{102} acitivity distributions over Hyderabad, India, based on the 20 April 1961 beta activity profile shown in Fig. 4 and the nuclide ratio data from the SMAC flights (Table 4), are presented in Table 6 and Fig. 5. The curve for Ce^{144} vs. altitude, Fig. 5, is constructed by assuming a Ce^{144} to total beta activity ratio of 0.34. The Ce^{144} values, Table 6, were obtained from the intercept of the altitude of each SMAC flight on the Ce^{144} curve, Fig. 5. Sr^{90} and Rh^{102} data, Table 6 and Fig. 5, are based on the Ce^{144}/Sr^{90} and Ce^{144}/Rh^{102} ratios for the SMAC flights, based on the data given in Table 4. The similarity in altitude trend for the Ce^{144} and Sr^{90} data of Fig. 5 indicate the approximately uniform composition of fission products vs. altitude which is reassuring with respect to the assumption used in constructing the Ce^{144} curve. The increase in Rh^{102}/Sr^{90} ratios vs. altitude is clearly apparent. These Sr^{90} and Rh^{102} concentrations at 60,000 to 70,000 ft., Fig. 5, are comparable to those obtained from U-2 aircraft samples at these altitudes over the equator. It is also of interest to note that the Rh^{102}/Sr^{90} ratios at the upper levels over Hyderabad, India, are about the same as their ratios for Direct-Flow unit samples from upper sampling levels at 32°N and 45°N latitude during the last half of 1960.

Discussion

The particle size measurements show that residual contamination at levels of 90,000 to 100,000 ft. and above resides in particles below 0.02 μ radius. This very low size range for radioactive aerosols has important implications with respect to methods of balloon and rocket sampling. The efficiency of IPC paper for particles of such small size is in doubt and probably is low, even for the design face-velocity of the Direct-Flow sampler currently employed by U.S. Atomic Energy Commission contractors in the upper-atmosphere monitoring program. At lower face velocities, which will result in the event of any obstruction to flow, the efficiency of fine particle collection on IPC paper will be further reduced. A filter pad of very fine fiber material, which provides for the efficient collection of particles in the mu range, is clearly required. Polystyrene microfiber material like the SRI filter used in the India flights or the commercially available Delbag-Microsorban 99/97, a filter of similar characteristics and manufactured by Delbag-Luftfiler (7), not only provide the requisite high efficiency, but have the additional advantage of low chemical and radiochemical impurity. The need for a larger area of filter material because of the higher resistance of these filters compared to IPC paper imposes no serious processing difficulties in view of the ease of destructive distillation of the polystyrene material.

TABLE 6

Radioactivity over Hyderabad, India, based on 20 April 1961 impactor-filter profile and SMAC sample nuclide ratio data

SMAC Flight	Altitude (10^3ft.)	Total beta[a] (G x cpm/10^3ft.3) (S.T.P.)	Ce^{144}[b] (dpm/10^3ft.3) (S.T.P.)	Sr^{90} (dpm/10^3ft.3) (S.T.P.)	Rh^{102}[c] (dpm/10^3ft.3) (S.T.P.)
2521	76.5	1210	410	93.2	66.5
2525	89.5	2800	945	210	< 418
2522	97.0	4500	1520	260	735
2534	68.5	700	236	73.5	31.8
2535	77.4	1290	435	75.4	42.5
2536	86.8	2330	787	226	~ 219
2537	97.3	4590	1550	468	~ 720

a. Based on impactor-filter profile of Flight 2542.

b. Ce^{144} taken as 0.34 x total beta.

c. Rh^{102} corrected to 12 August 1958.

The size distribution of radioactivity in the lower stratosphere appears to correspond with the peak in the number distribution and the lower end of the mass distribution of the natural sulfate aerosol. These results are consistent with those of Manson (8) who gives other evidence on the association of stratospheric radioactivity and the natural sulfate aerosol layer. It is indicated that the finely-divided radioactive aerosol mixes down from higher levels of the stratosphere and either seeds the formation of or becomes attached to sulfate aerosol particles. The nature and details of the mechanisms are not known. The experimental results clearly indicate that sedimentation must play a relatively minor role in the vertical transport of residual contamination, both within the lower stratosphere and across the tropopause.

The vertical distribution of radioactivity in the equatorial stratosphere shown in Fig. 6, must be taken as a tentative result because it is based on a single vertical profile of measurements. The increase in the concentration of Rh^{102} relative to fission products is clear on the point that the equatorial stratosphere is not well mixed but shows an increase in the relative contribution of high altitude sources with altitude over the range of observations. The marked increase in fission product concentration vs. altitude in this region and the uncertain efficiency of Direct-Flow unit data at upper levels indicate the need for more measurements of better quality over the balloon altitude range, with special attention to the equatorial region. Without such data, inventories of residual stratospheric contamination cannot be established.

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ATMOSPHERIC RADIOACTIVITY PATTERNS ALONG THE 80th MERIDIAN (WEST) 1959-1961

Luther B. Lockhart, Jr., Robert L. Patterson, Jr.
Allen W. Saunders, Jr., and Robert W. Black
U. S. Naval Research Laboratory
Washington, D. C.

Abstract

Some tentative conclusions based on the study of fission products and natural Pb^{210} in the air at ground level along the 80th meridian during the period 1959-1961 are presented. Seasonal variations in the quantities of fallout materials in the air of both hemispheres are documented. Changes in the relative concentrations of Ce^{144} and Sr^{90} during the past year indicate the apparent influx of younger debris into the lower atmosphere. A lack of seasonal variations in the ratio of these nuclides makes it difficult to accept this result as due to the increase contribution of Teak-Orange* debris.

Pb^{210} concentrations in the air along the 80th meridian show a latitudinal distribution similar to that of fission products, but with no systematic increase with altitude. The presence of Pb^{210} at the higher altitude sites is attributed to the vertical mixing of air masses containing Pb^{210} and/or Rn rather than gaseous diffusion of the latter material.

Introduction

The U. S. Naval Research Laboratory, (NRL) 80th meridian air-monitoring program, which is a cooperative endeavor financed by U. S. Naval Research and U. S. Atomic Energy Commission funds and supported by the U. S. Weather Bureau and various organizations outside the United States, has

* Very high altitude shots, Johnston Island, Operation HARDTACK I, 1958.

been in continuous operation since May 1956. The information obtained during this period has given some insight into the matter in which the air concentrations of fission product radioactivity at ground level are related to the intensity and geographical area of testing. Measurements made during the nearly 3-years moratorium on nuclear testing have enabled some conclusions to be drawn regarding the behavior of the various stratospheric sources of radioactive debris*. It is felt that there is sufficient information available from present and past records for a reliable estimate to be made of the future distribution of debris from the current series of tests in the U. S. S. R.

Procedure

As most of you are aware, this network consists of a series of 12 stations along the 80th meridian (west) which continuously sample the air at ground level using essentially constant volume blowers (positive displacement blowers) to draw about 1200 m^3/day of air through extremely efficient cellulose/asbestos filters (Army Chemical Corps type 6). Filters are currently changed at weekly intervals and returned to NRL for assay and radiochemical analysis. The individual samples are ashed, compressed to a pellet in a hydraulic press, and assayed for gross beta activity using standard techniques. Radiochemical analyses are performed on composite samples from each station for the nuclides Sr^{89}, Sr^{90}, Y^{91}, Cs^{137}, Ce^{141}, Pm^{147}, and Pb^{210}. During the period when W^{185} activity was available for measurement, it was substituted in the analysis scheme for Pb^{210}. All of the nuclides considered are beta emitters; the radioactivity is measured by use of low-level beta-counting techniques on counters which have been standardized against similar mounted samples of known disintegration rates.

Results

The radioactivity profiles along the 80th meridian for 1960 are shown in Fig. 1 as semilog plots of gross beta activity (dpm/m^3 of air) vs latitude. The common features of all these monthly profiles (and those of other years) are the maxima in the midlatitudes of each hemisphere and a minimum near the equator. The peaks at Miraflores, Panama Canal Zone, during 1960 are due to the French test of 13 February 1960 in the Sahara Desert. Several progressive changes are evident: (a) the Northern Hemisphere stations show a definite seasonal change with a maximum in the spring (independent of the peak associated with the French debris); (b) the tropical minimum shifts north of the equator during the period of the Northern Hemisphere fall minimum; (3) seasonal effects are not very apparent in the midlatitudes of the Southern Hemisphere; and (d) there is a sharper cut-off at the higher latitudes to the south as compared to the north, probably because of the lack of

*A list of recent publications are given at the end of this paper.

an appreciable burden of activity over the Antarctic stratosphere. During 1961 the gross activity levels have been about 50% lower in both hemispheres than in 1960.

The trends in the gross activity concentrations at Washington, Miami and Antofagasta are shown in Fig. 2. A definite periodicity is present at the Northern Hemisphere sites but not at Antofagasta, though a general upturn did take place during the Southern Hemisphere spring in 1960. The initial effect of the current U. S. S. R. series is apparent with an increase in fission product concentrations of nearly 2 orders of magnitude at Washington and Miami. None of this activity has yet been detected below the equator.

The effect of the French tests of February and April 1960 on the gross activity levels at four sites is shown in Fig. 3. Radiochemical analyses indicated that only trace quantities of fresh activity were collected at other 80th meridian sites. The contribution of Sr^{90} from the 13 February test to the total Sr^{90} in the air at the various sites during March 1960 is shown in Table 1. This test was a major contributor of Sr^{90} at only two sites and for this one month only. Traces of fresh activity were detected at a few sites during May 1960 but nowhere during July. This fast disappearance of the newer activity would indicate the absence of any measurable stratospheric component from this French test.

The French test of 1 April 1960 could not be distinguished above the general background of activity resulting both from the February test and from the normal spring rise in stratospheric deposition. The small test of 27 December 1960 was detected for a very short period and only at two sites, Miraflores, P.C.Z., and San Juan, P.R.; the test of 25 April 1961 was not detected either by gross beta measurements or by radiochemistry.

The same pronounced seasonal variation as was shown before in the air concentration of gross beta activity in the Northern Hemisphere is exhibited by the Sr^{90} in the air (Fig. 4). Actually the 1961 Sr^{90} spring peak is about 75-80% of that in 1960 while the gross beta peak of 1961 relative to 1960 was only 50%. By comparing the average Sr^{90} spring peaks in 1960 and 1961 relative to 1959, ratios of 0.148 and 0.126 are obtained, leading to apparent half-residence times for the average stratospheric source contributing to the troposphere of about 5 and 50 months, respectively, for the years 1959-1960 and 1960-1961. These values are significant in that they, perhaps, indicate a much shorter residence time for debris introduced into the Arctic than for that presently in the tropical stratosphere. However, if appreciable Teak and Orange debris were entering the troposphere during this period the effect would be the same as that observed. Corrections for an assumed 25% contribution from these high altitude shots would give a half-residence time of about 18 months for the older debris. It is unlikely that the Teak-Orange debris is this important; if so, its effect on the Southern Hemisphere should be relatively much greater.

Curiously, in both 1960 and 1961 the maximum Sr^{90} concentration was reached sooner at Miami and Thule than at Moosonee and Washington.

In the Southern Hemisphere there is a definite spring peak in the air concentration of Sr^{90} for the first time, clearly shown at Antofagasta and Santiago during 1960 (Fig. 5). No definite spring rise was noted at Puerto Montt or Punta Arenas. Gross beta measurements made at the South Pole during 1959-1961 do indicate seasonal variations there with maxima in their summer season (January-February).

Profiles of Sr^{90} along the 80th meridian during the periods of seasonal maxima and minima are shown in Fig. 6. For comparison the profile for January 1960 is shown to indicate the distribution during an intermediate period. It should be noted that the data for no one month will depict accurately the profiles during periods of maxima and iminma because of the different times of occurrence at the various sites. It is evident, however, that decided seasonal changes do occur which are roughly 6 months out of phase.

The ratios of the Ce^{144} to Sr^{90} activities in the air during the period 1959-1961 at the various sites outside the tropics are shown in Fig. 7. Following the spring peak of 1959, when the bulk of the Northern Hemisphere radioactivity came from the U. S. S. R. October series of 1958, the Ce^{144}/Sr^{90} activity ratio steadily decreased as the relative contribution of older debris increased. The top slanted line indicates the decay rate of Ce^{144}/Sr^{90} from a single source and extrapolates back to a zero time of 15 October 1959. The lower line serves to separate the ratios in the two hemispheres; it extrapolates to October 1957 for an initial Ce^{144}/Sr^{90} ratio of 44. During the spring of 1960 there was no identifiable influx of younger debris as might be expected if any quantity of the more recent U. S. S. R. debris remained in the atmosphere. In the Southern Hemisphere from mid-1959 through the end of 1960 the Ce^{144}/Sr^{90} ratio indicated much older debris and decayed at a fairly constant rate ($T_{\frac{1}{2}}$ = 293 days, approx.), though there is slight evidence of an influx of older debris in September-November 1959. The average age differential between hemispheres decreased from about 12 months to 2 months during an 18 month period. This effect cannot be explained by a transequatorial mixing process since there is no corresponding decrease in the age of debris in the Southern Hemisphere. Starting in mid-1960 in both hemispheres there has been a gradual decrease in the rate of decay of the Ce^{144}/Sr^{90} ratio as might be occasioned by the increasing influence of a source of debris younger than the prevailing average. This does not appear to be a seasonal effect as would be expected from an influx of Teak and Orange debris into the lower polar stratospheres. It may be due to another fairly young tropical source whose presence is just beginning to be felt; for example, debris from the United Kingdom test of September 1958 may have been introduced at a considerably higher altitude than the Operation HARDTACK debris and may just now have migrated to a position of influence on the tropospheric air. This is pure speculation, however.

The desirability of exploiting long-lived natural radioactive products as tracers for atmospheric processes has been long recognized, particularly as the fission product concentrations have become lower and more difficult to measure. Furthermore, such products have a different source than do fission products and a knowledge of their behavior will supplement the information obtained from the more routine fission product measurements. Consequently, part of the radiochemical effort has been expended in determining the contribution of the long-lived natural radioisotope Pb^{210} to the total activity of the air. It has been found that Pb^{210} for the past year has been a major contributor to the measured gross beta activity at ground level, generally contributing between 10% and 40% of the total. This has greatly increased the difficulty in interpreting results of gross beta measurements.

The data obtained during the past year indicate that at ground level the Pb^{210} concentration at a given site can vary widely from month to month. There is as yet insufficient data to document seasonal changes in the Pb^{210} concentrations; however, as indicated in Fig. 8, there is a definite latitude dependence, with the pattern of Pb^{210} concentrations resembling those of Sr^{90} along the 80th meridian. The unsymmetrical shape is due in part to the fact that Pb^{210} is derived from Rn which, like Sr^{90}, has its major source in the Northern Hemisphere. The tropical minimum is perhaps related to the rainfall patterns there. The high altitude sites of Mauna Loa (3394 m), Chacaltaya (5220 m) and the Amundsen-Scott station at the South Pole (2800 m) all have Pb^{210} concentrations (activity per unit mass of air) not greatly different from those of sea-level sites at similar latitudes. This would indicate that the Pb^{210} encountered at high altitudes is the result of vertical mixing of air masses from below, rather than of gaseous diffusion of the Rn parent and also that no active removal processes are operating at high altitudes. A study of the gradient of Pb^{210} across the tropopause and in the stratosphere should give an indication of the porosity of this "barrier" and indicate where the holes are located. Such a study might give evidence as to whether air masses are exchanged locally or whether there is a preferential place of influx into the stratosphere quite distant from the areas where Sr^{90} leakage occurs.

Summary

The information presented here covers some of the highlights of the work during the past year. Much detail has of necessity been omitted. Among the more important conclusions derived from this and similar work elsewhere is that seasonal maxima in stratospheric deposition do occur in both the Northern and Southern Hemispheres and are essentially 6 months out of phase; consequently, similar meteorological processes involving the seasonal variation in the mixing of stratospheric and tropospheric air are operating in both hemispheres.

The size of the available stratospheric source, as estimated from ground level measurements, apparently has not changed considerably during the past year. It is yet not certain whether this is entirely due to the recognized much longer residence time of debris in the tropical stratosphere relative to the emperate or polar stratosphere, or whether it is due in part to an influx of material from high altitude shots whose influence is just beginning to be felt at ground level. It is considered that this fact must be determined from measurements of the Rh^{102} nuclide introduced with high-altitude HARDTACK debris, since variations in fission product ratios are subject to a variety of interpretations. If it is indeed true that the Teak and Orange or other high altitude shots have as yet made no great contribution to the activity at ground level, then it is evident that transequatorial migration of debris in the stratosphere must have taken place sufficiently to override the loss due to deposition in the Southern Hemisphere. It may be concluded, in any case, that the rate of transequatorial mixing in the stratosphere is slow.

The interpretation of results of future measurements in the Northern Hemisphere will be extremely complicated for a while as the result of the current U. S. S. R. tests. However, the great quantity of young debris in the Northern Hemisphere provides some extremely sensitive tracers for detecting the transequatorial mixing or migration of air masses. The few reported indications of U. S. S. R. debris appearing in the Southern Hemisphere have been inconclusive because of the relatively little difference in the composition of the background activity in the two hemispheres. At the present time this will not be a limitation.

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NOTE: NRL reports are issued by U. S. Naval Research Laboratory, Washington.

TABLE 1

Contribution of the French Test of 13 February 1960 to the Total Sr^{90} in the Air at Various Sites During March 1960

Site	Activity (dpm/100 Std. m^3)			Contribution of French Test
	Sr^{89}*	Sr^{90} equivalent**	Sr^{90} Total	%
Thule	0.26	0.0016	0.908	0.18
Moosonee	0.56	0.0034	0.856	0.40
Washington	0.65	0.0039	0.940	0.41
Miami	7.3	0.044	1.51	2.9
Mauna Loa	21.6	0.130	1.39	9.3
Miraflores	78.0	0.470	0.858	55.
Guayaquil	15.7	0.095	0.170	56.
Lima	0.34	0.0021	0.115	1.8
Chacaltaya	0.26	0.0016	0.131	1.2
Antofagasta	0.30	0.0018	0.303	0.6
Santiago	0.0	0.000	0.360	0.0
Puerto Montt	0.0	0.000	0.311	0.0
Punta Arenas	0.0	0.000	0.163	0.0

* Corrected for decay to 13 February 1960
** Based on Sr^{89}/Sr^{90} ratio of 166 at time of fission

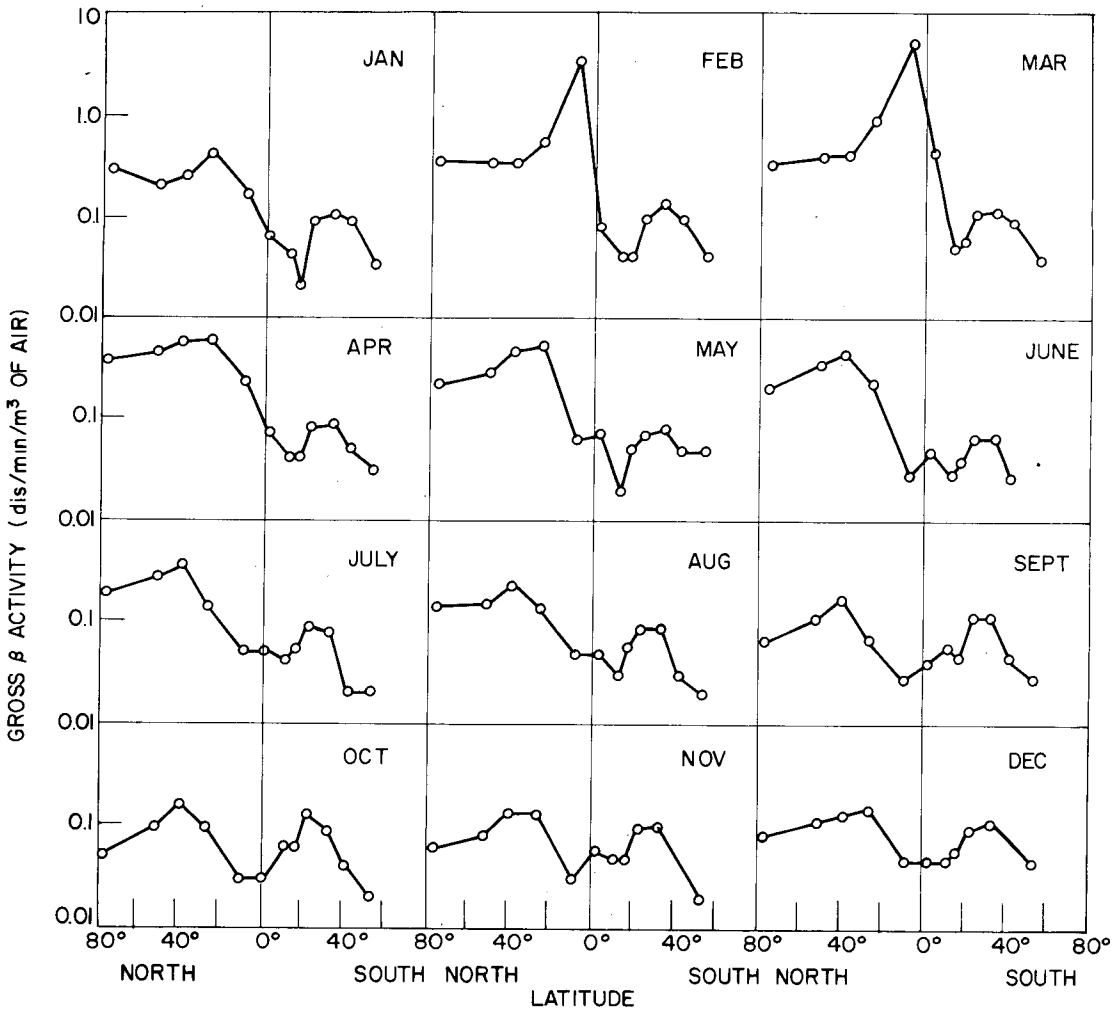

FIGURE 1. MONTHLY PROFILES OF GROSS β ACTIVITY IN THE AIR AT GROUND LEVEL ALONG THE 80TH MERIDIAN DURING 1960

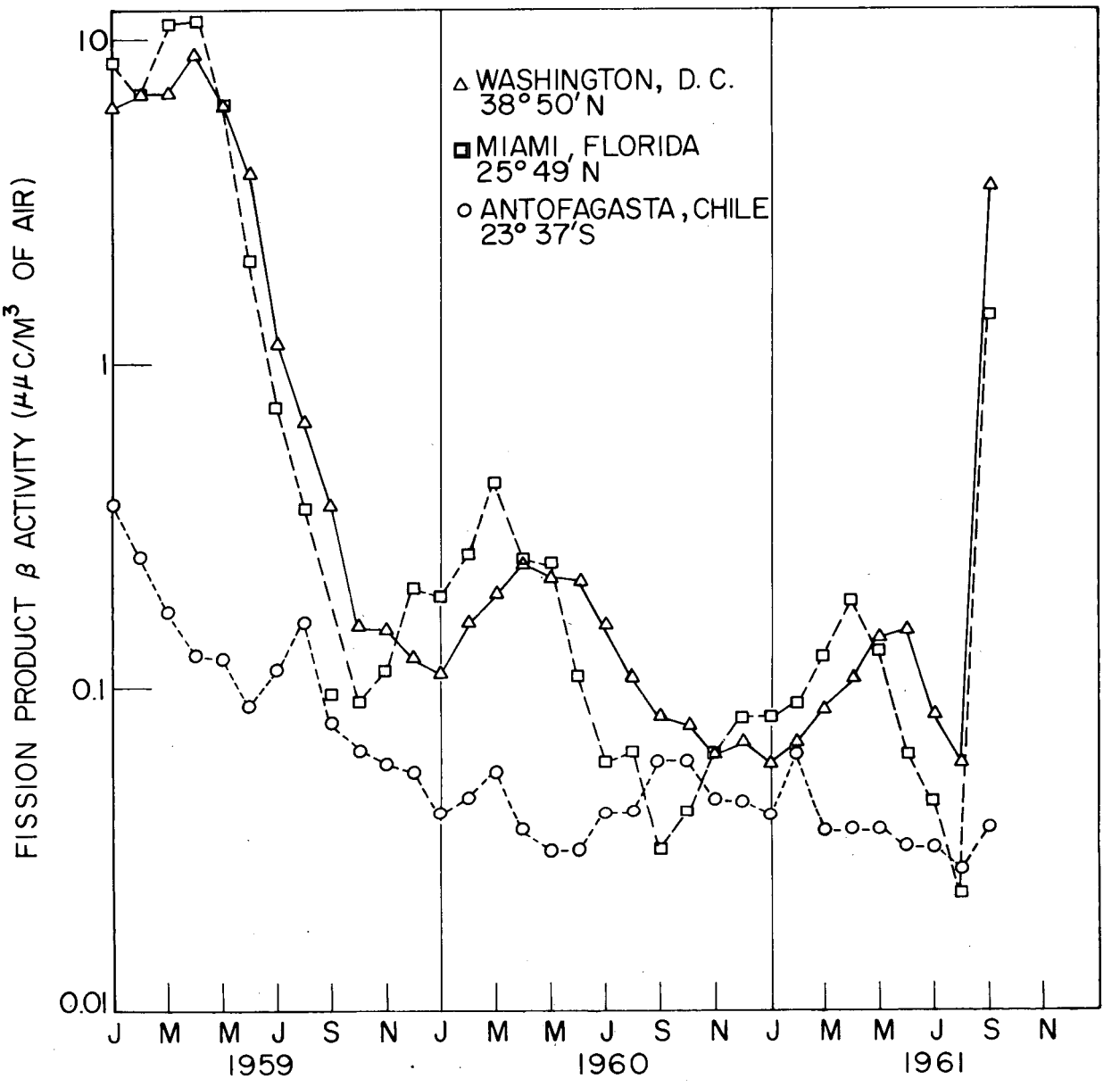

FIGURE 2. GROSS FISSION PRODUCTS IN THE AIR

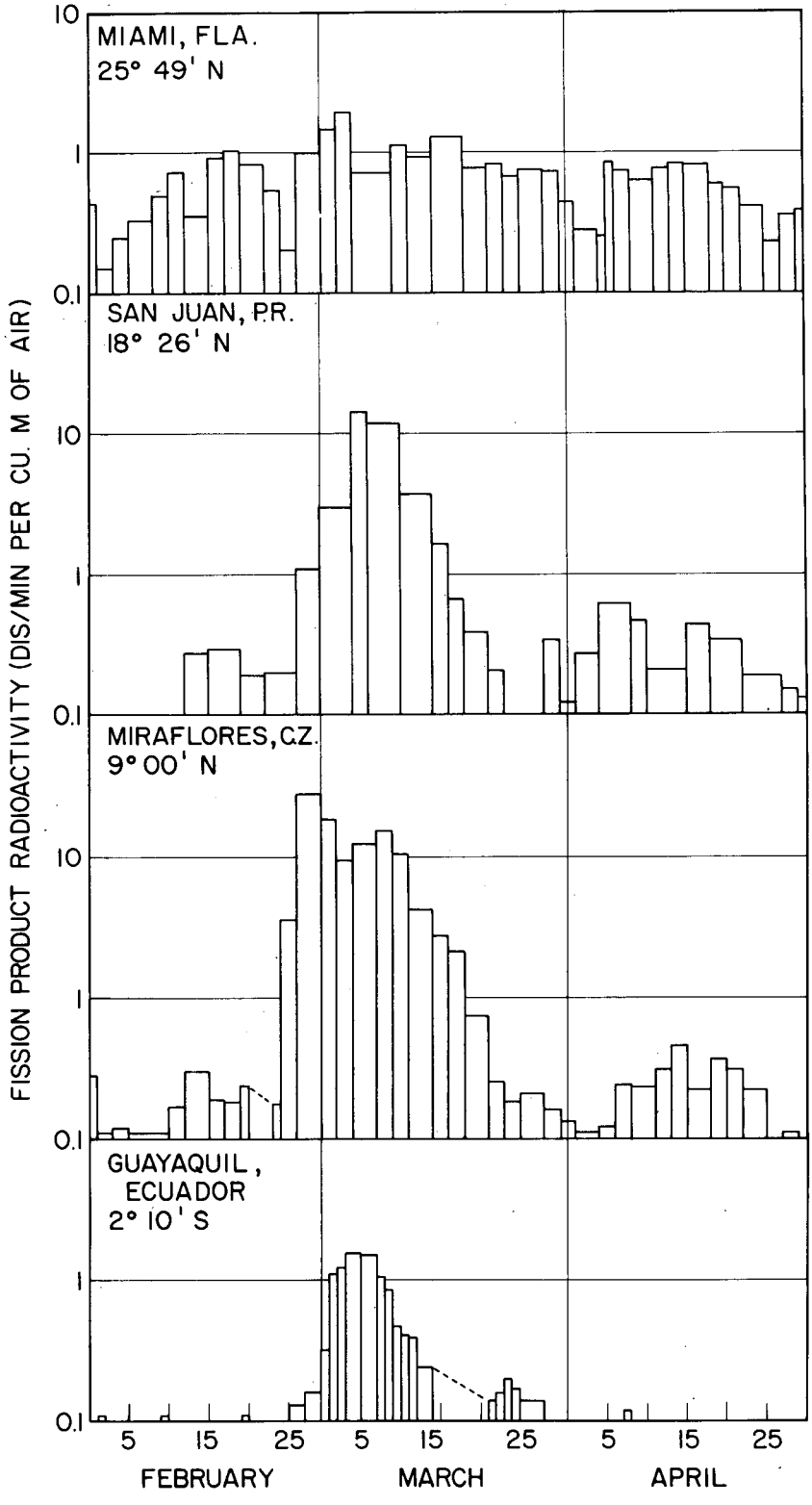

FIGURE 3. CHANGES IN THE CONCENTRATION OF FISSION PRODUCTS RADIOACTIVITY IN THE AIR ALONG THE 80TH MERIDIAN (WEST) FOLLOWING THE FRENCH NUCLEAR TESTS OF 1960

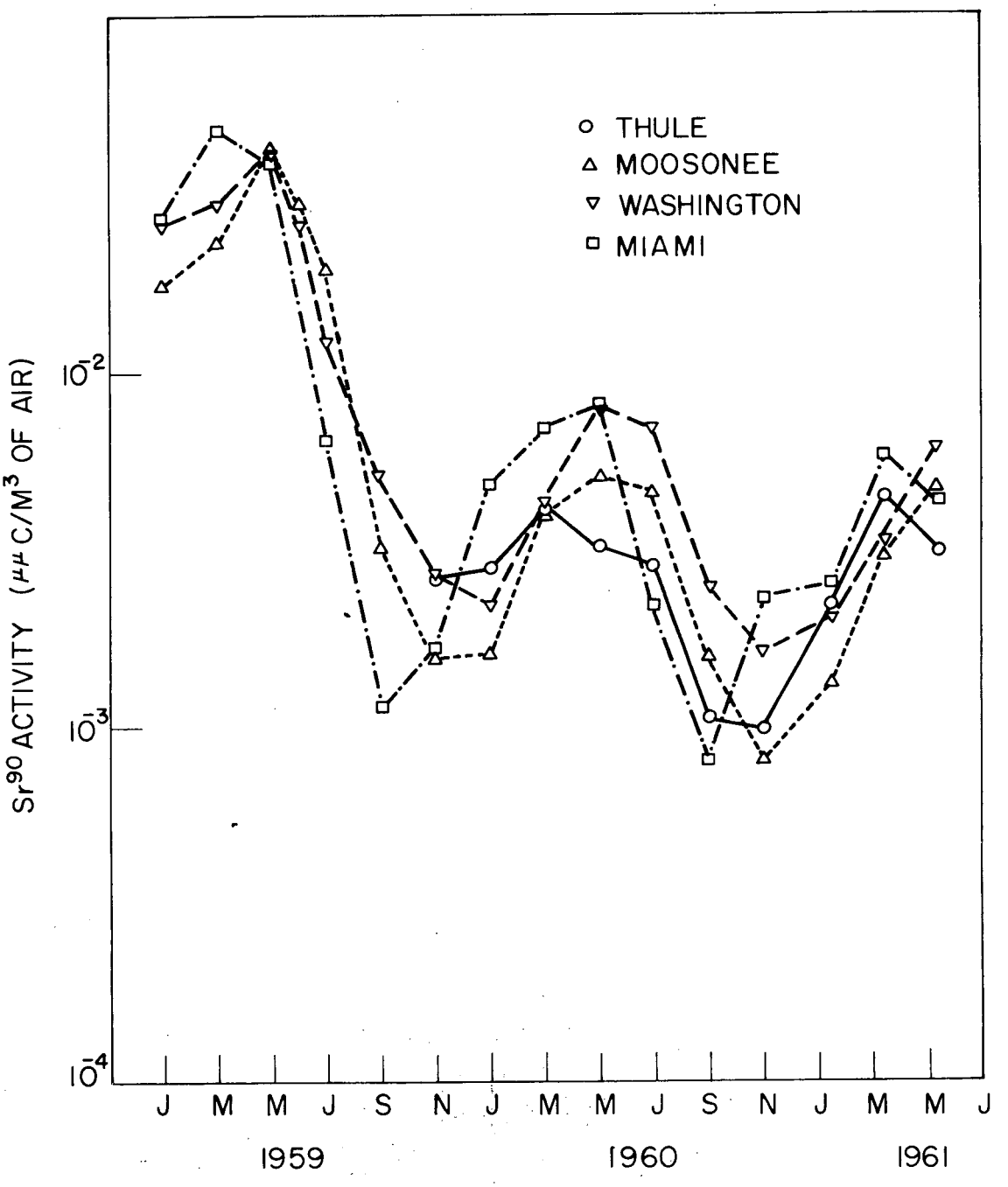

FIGURE 4. CONCENTRATION OF Sr^{90} INTHE AIR OF THE NORTH TEMPERATE ZONE 1959-1961

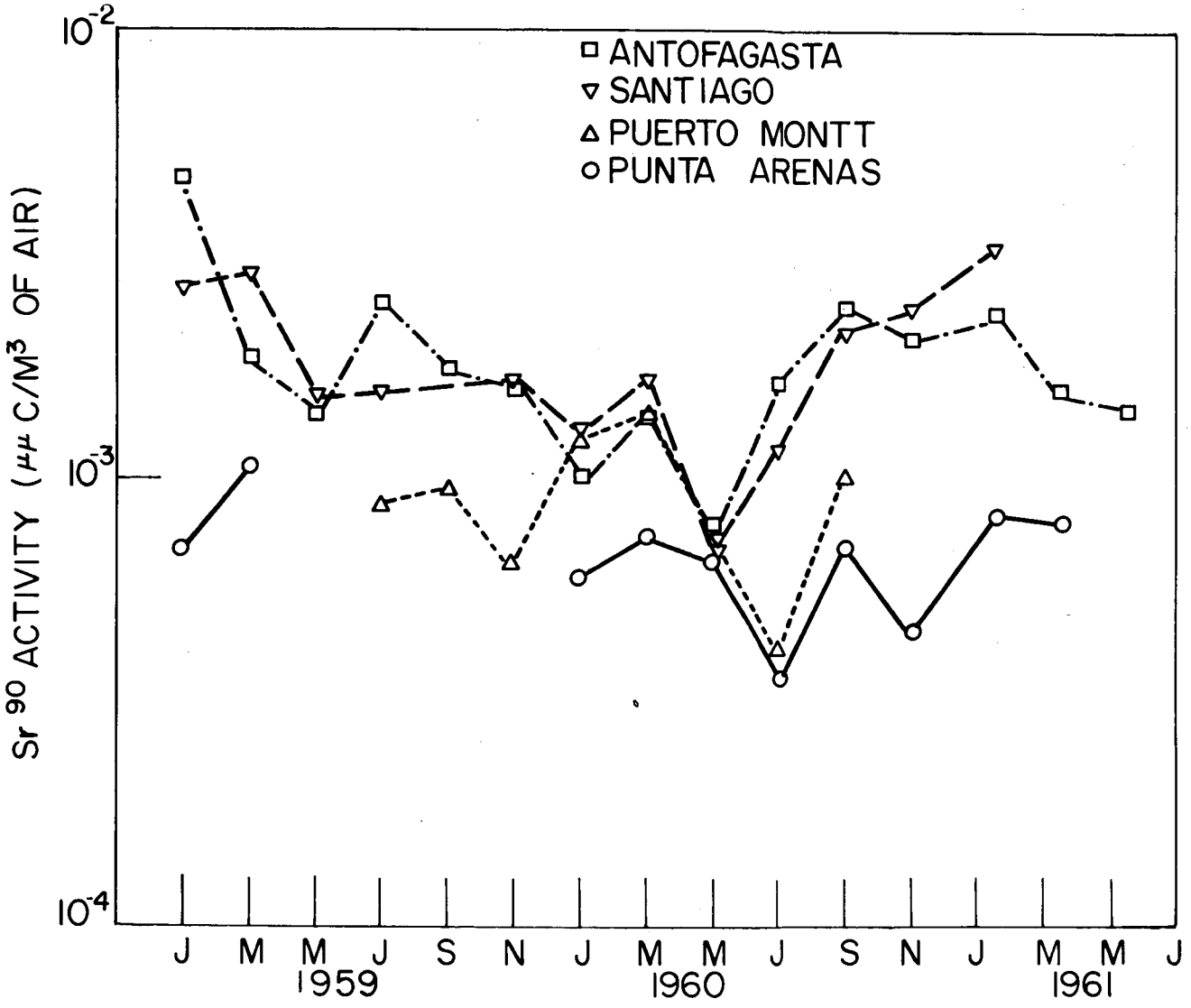

FIGURE 5. CONCENTRATION OF Sr^{90} IN THE AIR OF THE SOUTH TEMPERATE ZONE 1959-1961

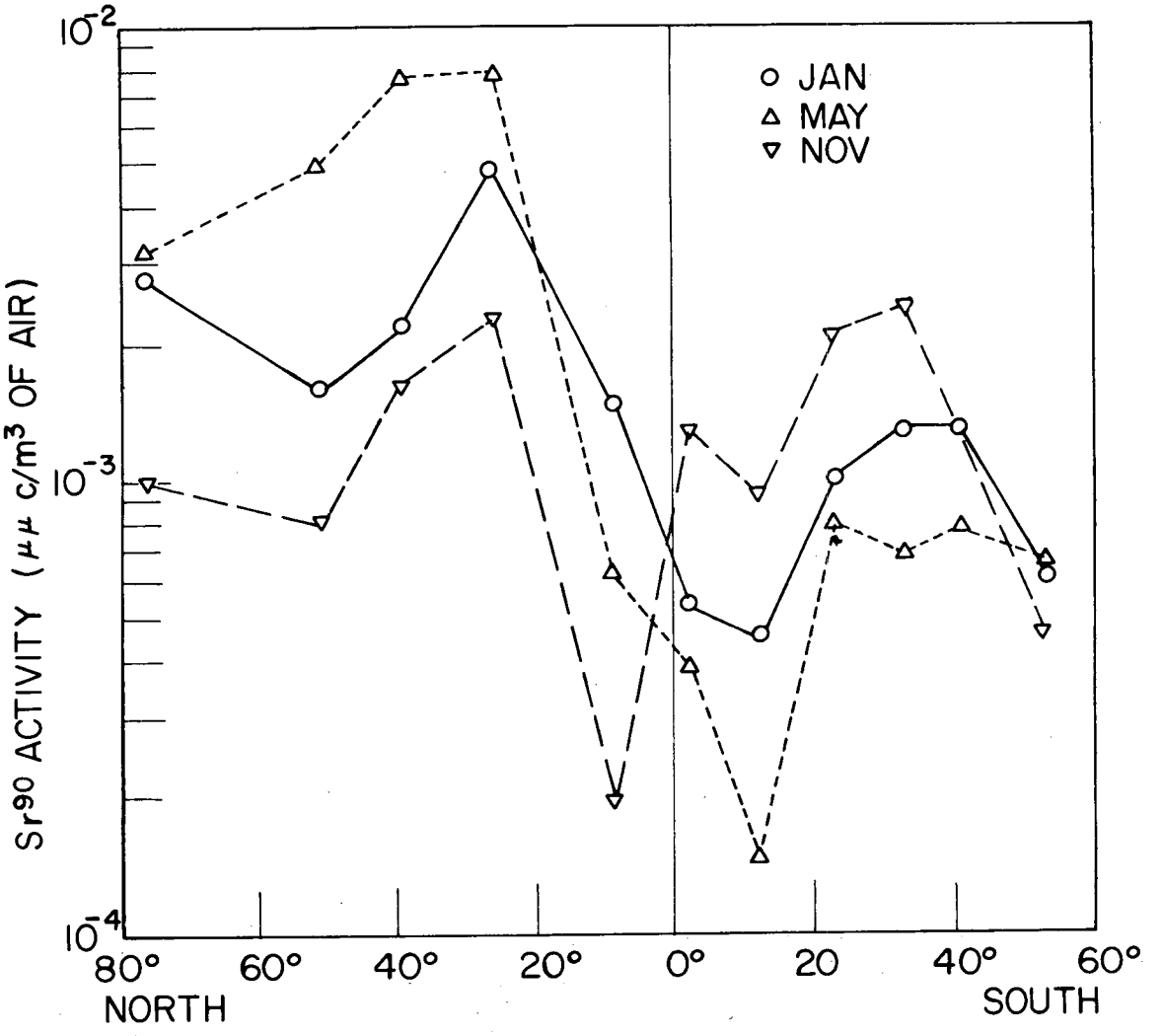

FIGURE 6. PROFILES OF Sr^{90} ACTIVITY ALONG THE 80TH MERIDIAN DURING 1960

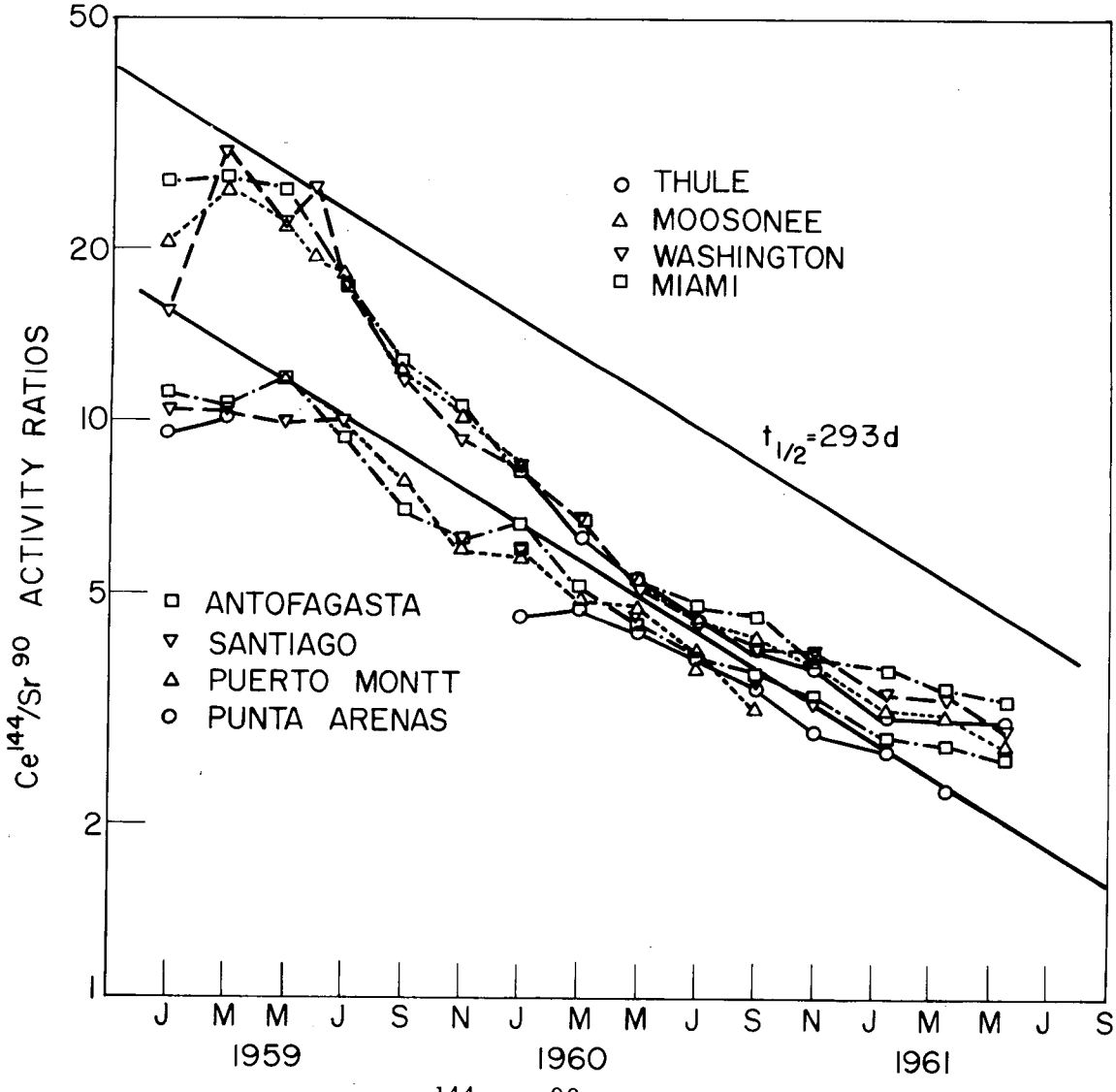

FIGURE 7. RATIOS OF Ce^{144} to Sr^{90} ACTIVITY AT VARIOUS SITES DURING 1959- 1961

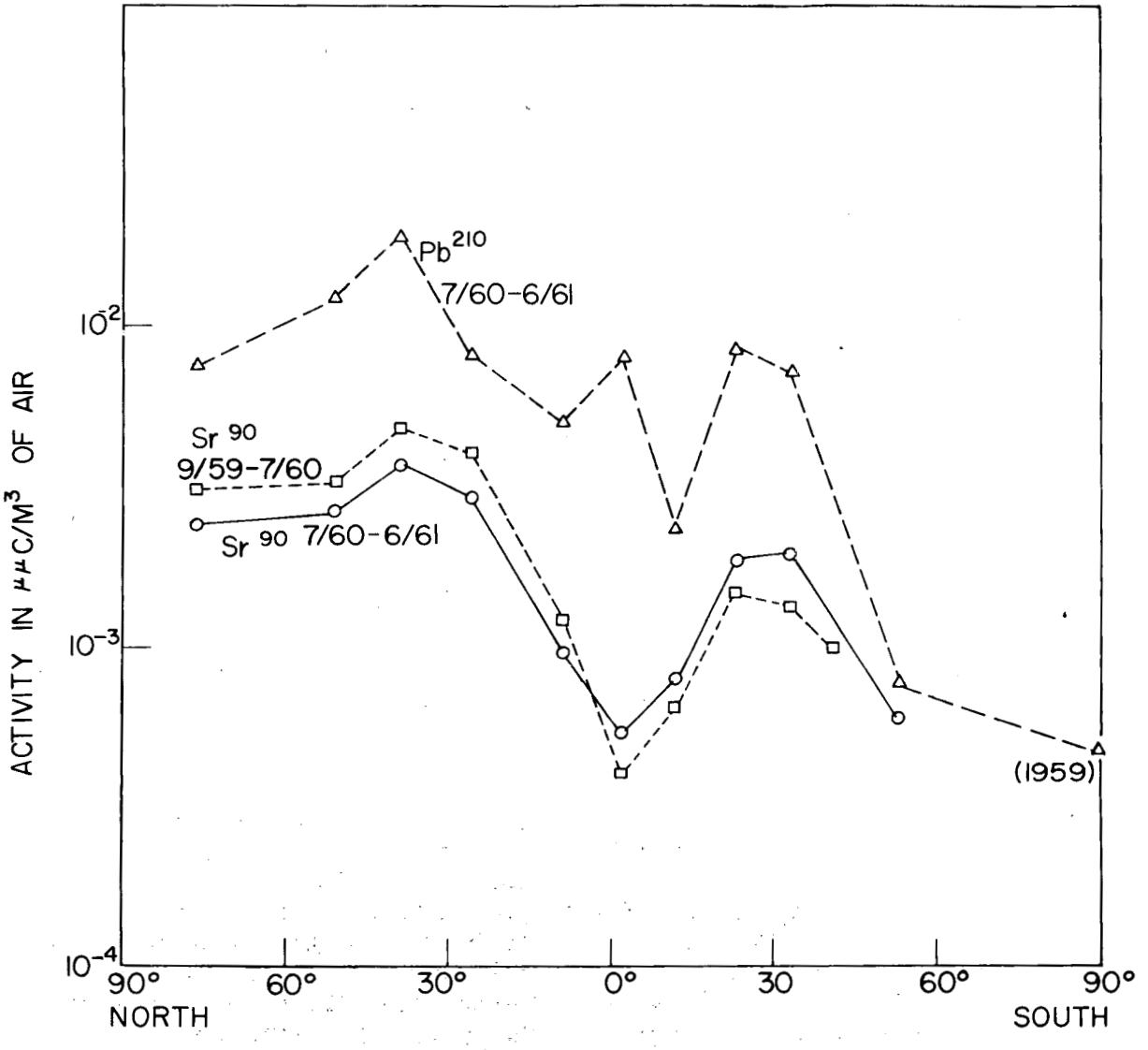

FIGURE 8. PROFILES OF THE YEARLY AVERAGES OF Sr^{90} and Pb^{210} IN THE AIR AT GROUND LEVEL ALONG THE 80TH MERIDIAN

GENERAL CIRCULATION OF THE STRATOSPHERE

Arnold A. Barnes, Jr.
Massachusetts Institute of Technology
Cambridge, Massachusetts

Abstract

Using data from 212 stations over the Northern Hemisphere, the meridonal circulation at 50 mb for the period July through December 1957 was obtained. The mean meridional circulation was also computed from the adiabatic assumption. The mean adiabatic and diabatic motions have the same magnitures but generally oppose each other.

Both the mean circulation and the eddies must be considered in any model of long term stratospheric transport of radioactive material.

Since the atmosphere is its own best model we have used the actual observations to obtain the mean south to north velocities at 50 mb for the period of July 1961 through 31 December 1961. Fig. 1 shows the 212 stations covering the Northern Hemisphere which were used in our study. Observations at times 00Z and 12Z at the 100 mb, 50 mb, 30 mb and 10 mb levels were used. The adiabatic vertical motions were computed for the layer 100 to 50 mb and 50 to 30 mb. 24-hour time differences were used to reduce the errors due to the sun shining on the instruments.

Three-month time means of these vertical motions, $\frac{dp}{dt}$, were computed for each of the stations and plotted on maps. These maps were analyzed and grid point values were taken at each 10° of longitude and every 5° of latitude from 80° N to the equator. Fig. 2 is a typical map of the adiabatic vertical motion. Maximum values correspond to about 1/2 cm/sec.

Using the continuity equation, it was possible to obtain the adiabatic horizontal motions along the pressure surface. Fig. 3 shows the mean

adiabatic meridional circulation for the period July through September 1958. Notice the close agreement between the 00Z and 12Z circulations.

The first column of Table 1 lists the latitudinal means of the 6-month time averages of the southerly components of the actual winds at 50 mb. Six-month averages were necessary to separate the signal from the noise. The second column lists the average 50 mb southerly components computed from the adiabatic vertical motions. Values south of 20° N are in doubt because of the small number of stations in the eqatorial region and because of the errors introduced at low latitudes by using the thermal wind relation in computing the adiabatic vertical motion. The third column is obtained by subtracting the second from the first. These values may be interpreted as the mean horizontal motions due to diabatic effects in the lower stratosphere. Notice that these mean horizontal diabatic motions are in the samedirection as those obtained by Murgatroyd and Singleton (1), but are much smaller in magnitude.

The adiabatic and diabatic motions are of comparable size bu usually have opposite directions.

Jensen's work (2) indicates that there is a change in the meridional circulation from Massachusetts Institute of Technology for the complete year 1958 using the 212 stations shown above.

Concluding we would say that for the lower stratosphere:

1. The diabatic heating is very important and cannot be disregarded.

2. Using our method, over 3 months of actual wind data must be used to separate the mean vertical components from the noise.

Newell has used these actual mean winds in computing transports in the stratosphere, and, as he shows in the following paper (page) the mean meridional motions cannot be disregard in computing stratospheric transports.

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TABLE 1

Six-month horizontal meridional winds at 50 mb
(cm/sec. Positive values indicate northward motion)

Actual	Adiabatic	Diabatic	Latitude
2	-7	9	80° N
-26	-9	-17	75
-5	-8	3	70
6	-7	13	65
6	-7	13	60
1	-12	13	55
-11	-15	4	50
-15	-17	3	45
-17	-18	1	40
-14	-17	3	35
-8	-15	7	30
-2	-13	11	25
5	-9	14	20
12	-4	16	15
10	0	10	10
6	3	3	5° N
2	4	-2	0°

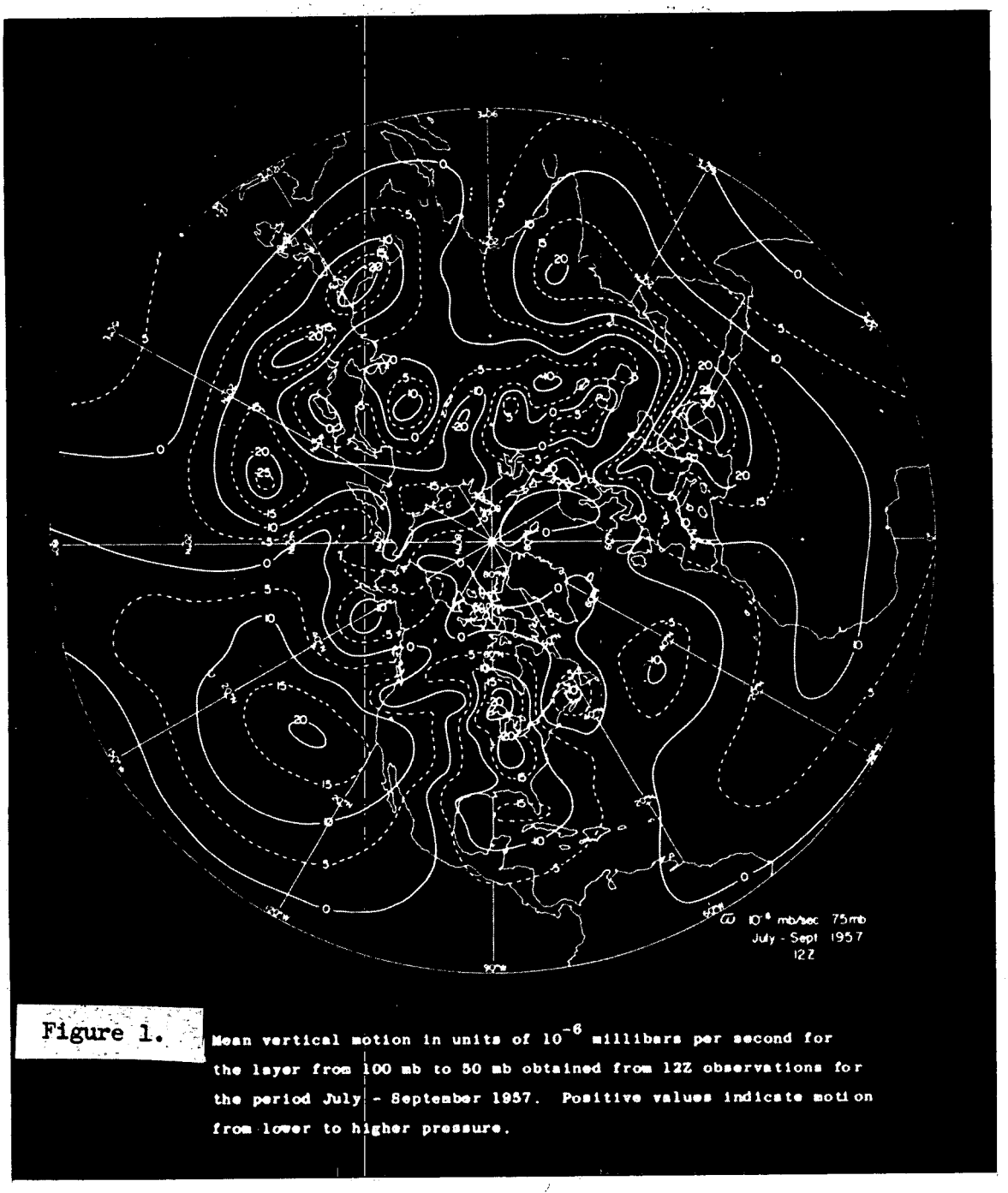

Figure 1. Mean vertical motion in units of 10^{-6} millibars per second for the layer from 100 mb to 50 mb obtained from 12Z observations for the period July - September 1957. Positive values indicate motion from lower to higher pressure.

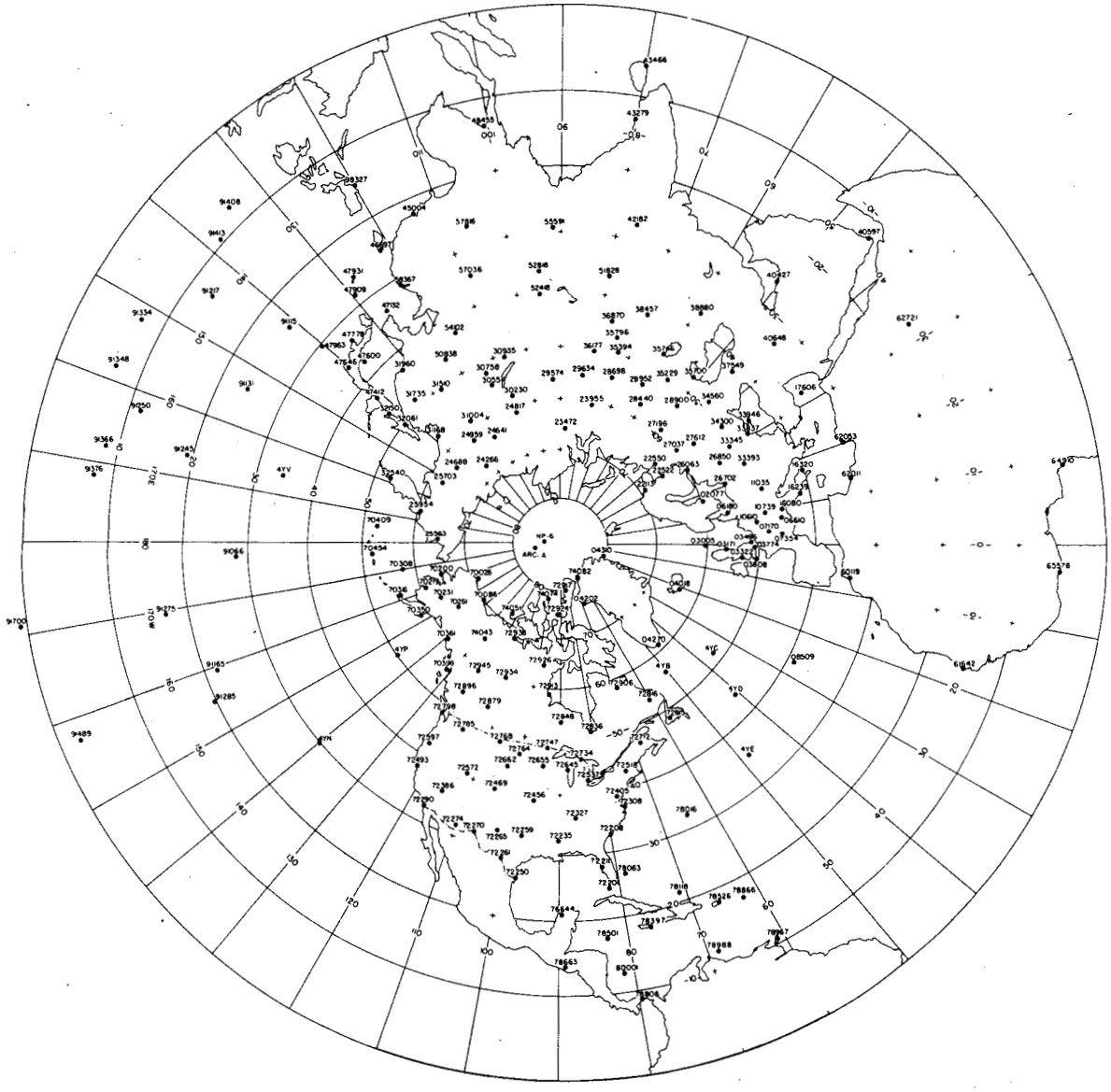

Figure 2. Stations, by international index number, used as source for stratospheric data.

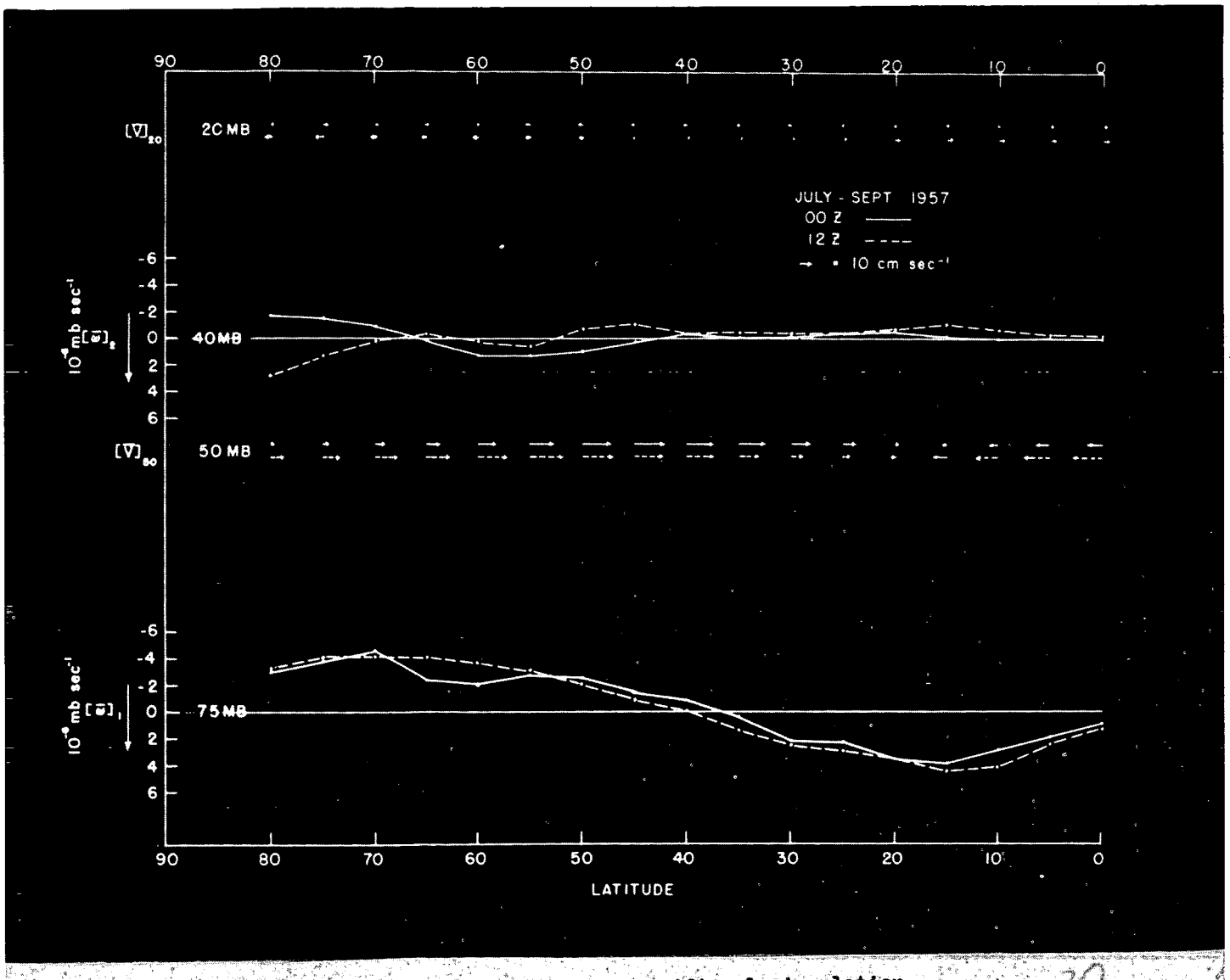

Figure 3. Mean adiabatic meridional circulation, July through September 1958

THE TRANSPORT OF OZONE AND RADIOACTIVITY IN THE ATMOSPHERE: IMPLICATIONS OF RECENT STRATOSPHERIC CIRCULATION FINDINGS.*

Reginald E. Newell
Department of Meteorology
Massachusetts Institute of Technology
Cambridge, Massachusetts

Background Note

The Planetary Circulations Project, directed by V.P. Starr, is presently investigating the circulation of the stratosphere using the method of attack that has already been extensively applied to the troposphere (see for example Starr (1, 2)). From the actual wind and temperature observations at 100, 50, 30 and 10 mb calculations are being made of the average wind and temperature field and the vertical and horizontal transports of angular momentum, kinetic energy, potential energy and heat. One of the goals is an understanding of the mechanisms of transfer within the stratosphere and this will be a prerequisite to any actual "explanation" of the observed behaviour comparable to that which now exists for the troposphere.

In the present paper we shall treat one particular aspect of the work that seems pertinent to the conference theme, rather than attempting to make a comprehensive survey of all the work being performed.

Introduction

Two problems of meteorological concern that have been raised in connection with the high-yield nuclear weapons' tests are: Why is it that the maximum tropospheric concentrations of fission products are found in middle latitudes whereas the debris is introduced into the stratosphere at low and high latitudes? Why are these tropospheric concentrations greatest in the spring season?

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Let us assume for present purposes that a large fraction of the tropospheric-stratospheric mass exchange occurs in the vicinity of the tropopause gap or the baroclinic zone of middle latitudes. Such transfer has been suggested by several authors and there is some support from observations (see for example Ramanathan (3), Machta (4), Brewer (5), Newell (6,7), Staley (8) and Roach (9)). Brewer presented some actual ozone soundings made with his chemical sonde which showed layers of relatively ozone-free air in the lower stratosphere of middle latitudes; he interpreted these as possibly due to isentropic transfer from the tropical troposphere. Roach's measurements, also with Brewer's apparatus, show an association in the region of the tropopause gap and jet stream between ozone rich layers and water-vapour poor layers. Again the data suggest that the ozone rich air in the troposphere in this vicinity has entered via the gap). With this assumption the original questions could be modified to read: How is transport accomplished <u>within</u> the stratosphere from low and high latitudes to middle latitudes? Are the spring maxima due to larger transports within the stratosphere in late winter or to increased stratospheric-tropospheric exchange or to both causes?

Similar questions have long puzzled meteorologists interested in atmospheric ozone. The photochemical equilibrium theory of ozone predicts maximum amounts in the summer and at low latitudes yet observations of total ozone show that maximum amounts occur in the late winter and spring at high latitudes. Below about 25 km ozone is not in photochemical equilibrium; in fact it can be treated as a conservative tracer as it is effectively shielded from ultra violet radiation by the ozone and oxygen above. The theory and observations can be reconciled if one postulates a northward transport of ozone in the lower stratosphere by a process that is more efficient in the winter. Brewer (10) and Dobson (11) suggested that the transport mechanism was a mean meridional motion. Martin (12) presented some calculations of the ozone flux which showed the importance of quasi-horizontal eddy mixing in the lower stratosphere. Herein we shall present some further transport calculations in which both eddy diffusion and the mean motions discussed in the preceeding paper (page 204) by A. A. Barnes, Jr., are taken into account.

We should emphasize here that the ozone is not of purely academic interest at this meeting; it has a stratospheric source, is subject to stratospheric-tropospheric exchange, and has a maximum concentration in both the middle latitude tratosphere and the lower troposphere in late winter or spring (for the latter fact I am indebted to C. E. Junge)

Computations of Ozone Transport

Suppose that the ozone concentration at a particular point at a given time is O ; we can write

$$O = \overline{O} + O'$$

where the bar represents the time mean and the prime the departure from the mean.

Likewise the northward component of the wind V may be written

$$V = \overline{V} + V'$$

The instantaneous northward transport of ozone will be given by

$$OV = \overline{O}\,\overline{V} + O'V' + O'\overline{V} + \overline{O}\,V'$$

and the time averaged transport is

$$\overline{OV} = \overline{O}\,\overline{V} + \overline{O'V'}$$

The equation is an expression of the fact that the northward transport of ozone is due to transport by mean meridional motions ($\overline{O}\ \overline{V}$) and transport by transient eddy processes. For the latter term to be effective in the present problem it is necessary that northward moving parcels of air contain more ozone than southward moving parcels.

In addition to the transporting agencies of transient eddies and mean motions there are the standing eddies. If there is a systematic variation of amount of ozone with longitude which is related to large-scale semi-permanent troughs and ridges then there may be a northward transport on this account.

Measurements of the concentration of ozone in the stratosphere are not available on a synoptic basis but there is a good correlation between the ozone concentration in the lower stratosphere (12-24 km in middle latitudes) and the total ozone amount in a vertical column (3, 13). Total ozone amounts are available on a synoptic basis; we used those taken at 25 stations in the northern hemisphere during the International Geophysical Year (IGY). The stations are listed in Table 1; where the wind site was not at the ozone station the wind site is listed in parentheses. Calculations were made of

$\overline{O}$, $\overline{V}$, $\overline{O'V'}$; the standard derivations $\sigma(o)$ and $\sigma(V)$; and the correlation coefficient r (0,V) . Both 50 mb and 100 mb winds were used at the times 0001 and 1200 GMT. Many stations report a series of ozone measurements throughout the day; in these cases the ozone amount closest to the wind time was selected. Otherwise the single ozone value reported was used with both winds. (Similar computations have been made for the westerly component of the wind but will not be discussed in this paper).

A summary of the transient eddy flux data is shown in Table 2. The Japanese stations have been placed in a separate group because of their vicinity to the very strong jet stream in that area as shown by Barnes' wind data. The few stations at high latitudes are also shown separately. The middle latitude stations, separated into three latitude belts, show in general a northward flux of ozone with the largest values occurring in the January-March period. The implication is of course that the stratospheric quasi-horizontal eddy mixing processes are more vigourous in this period. By the fall the eddy mixing is less by a factor of three or four and the region of maximum transport has moved towards the north. The eddy transfers calculated are from the northward component of the wind. In order for there to be a transfer away from the photochemical equilibrium region and therefore down the ozone concentration gradient, northward moving parcels should be descending and southward moving parcels should be rising. There is some indirect evidence in favour of this hypothesis in the observations of a counter-gradient heat flux by transient eddies at 200 and 100 mb in middle latitudes reported by White (14). They could be interpreted as due to northward moving parcels subsiding and warming adiabatically.

Some ozone budget calculations are shown in Table 3. As a guide to the importance of the various terms the actual observed change in ozone content of the polar cap north of 40° and 50° N has been calculated. In the spring indications are that the transient eddy flux is sufficient to account for the observed increase. The standing eddy term cannot be determined properly until data from many more ozone stations are used. Thus far there are no measurements of the mean meridional circulation in the spring. Barnes' measurements which are an average for the period July-December 1957 have been included to give a representation of the flux in the July-September periods. Transient eddy effects are subordinate to the effects of mean meridional motions in this period. We hope that the mean motions from the next 6 months of the IGY data will be available in the next few months. One value is available for the downward flux in the troposphere (15); it has been converted into an equivalent flux over one quarter of the globe and is shown in Table 3. Notice that the content change in the fall could be due entirely to loss

to the troposphere. Many more values of the tropospheric flux would be needed to establish this point. Junge has recently urged the establishment of a meridional network of stations which would report surface ozone concentrations (similar to that employed by Lockhart (see page 188)).

Comparison with Other Evidence

a) W^{185} measurements, reported by Feely and Spar (16) and kindly provided to the author by A. K. Stebbins, III (Defense Atomic Support Agency), indicate a northward flux by eddy processes down the concentration gradient and to a certain extent along the isentropic surfaces (Figs. 1 and 2). The apparent cross-isentropic flow may be partly due to diabatic effects and partly to the fact that mean isentropes are used. Note that the maxima in the two hemispheres are on the same isentrope and that the maxima remain in its vicinity from one period to the next (although the periods shown here are not strictly comparable). The isentropes used here are taken from the work of Taylor (17) and refer to 170° E. We do not yet have available from our own work results from the southern hemisphere. The isentropes from Barnes work for 80° W have been used in a similar superposition that we reported earlier (7); the same general features are shown by both sets. In fact, in detail too there is some agreement as the secondary maxima appear to move downwards by about 12 degrees in potential temperature. Some of the change in the Northern Hemisphere could have been due to gravitational settling.

b) The configuration of the isentropes and tungsten isolines suggests that northward moving parcels are sinking and southward moving parcels are rising in the stratosphere of 10 - 50° N at these longitudes. This is just the motion required by the ozone flux as already discussed.

c) The concept of northward moving parcels descending and therefore warming adiabatically provides an explanation for the counter-gradient flux of heat by eddy processes at 100 mb reported by White (14).

d) Loisel and Molla, until recently two of my colleagues on the Planetary Circulations Project, have shown (18) that the covariance $\overline{V' W'}$ is negative in the lower stratosphere south of 50° N and positive to the north. (W represents the vertical component of the wind). Again the interpretation is that northward moving parcels are sinking.

e) Some preliminary studies have been made of the angular momentum budget of the region by Barnes. Indications are that transient eddy effects transport angular momentum northward in winter at 50 mb in the region south of 60° N, and transport it southward north of this latitude. Again the shuffling process is more important in winter.

f) Martin and Brewer (19) showed from a study of trajectories that particles which had moved from the south over Europe contained more ozone and had higher temperatures than those which moved from the north at 100 mb.

Concluding Remarks

Both quasi-horizontal eddy mixing and mean meridional motions must be taken into account in attempts to explain the movement of trace substances in the stratosphere. The ozone data indicate more quasi-horizontal eddy mixing in the winter and this provides a possible explanation for the increased concentration of ozone in the middle and high latitude lower stratosphere (which is followed incidentally by increases in the tropospheric concentrations of both ozone and fission products). The quasi-horizontal eddy flux is of the right order of magnitude to account for the increase. In the fall both types of transfer are effective; the observed decrease in ozone content could be due either to southward transport by the mean motion or to loss into the troposphere. Many more calculations will be required before the role of the standing eddies can be determined.

If we had asked the two opening questions with reference to ozone it would seem reasonable to answer that transfer within the stratosphere is accomplished principally by quasi-horizontal eddy motions in the winter and that the spring maximum was a result of these processes introducing more ozone in winter than in summer into the lower middle and high latitude stratosphere. From this region it appears that escape occurs into the troposphere and regardless of the actual stratosphere-tropospheric exchange process there will be a maximum exchange in late winter if the transfer occurs down the concentration gradient.

The case of fission products is not exactly analogous to that of ozone as there are two sources of fission products, at high and low latitudes in the stratosphere. It is of interest, however, that Barnes' angular momentum flux data show a greater eddy transfer in the winter than in the summer between high and middle latitudes. Presumably this transport would result in the high latitude debris reaching regions of the middle latitude baroclinic zone favourable to stratospheric-tropospheric exchange.

The ozone flux data suggest that the pattern of events is not exactly the same from year to year. For example the Icelandic station shows a southward flux in October-December 1958 almost equal to that in January-March 1958. One must therefore exercise considerable caution in any attempts to forecast the middle-high latitude exchange intensity. Steps are being taken to extend this type of study over a period of years.

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TABLE I

Ozone Stations

Station	International Index No.	Location	
Marcus Island, Pacific	91-131	24°17'N	153°58'E
Torishima, Japan	47-963	30°29'N	140°18'E
Tateno, Japan	47-646	36°03'N	140°08'E
Sapporo, Japan	47-412	43°03'N	141°20'E
Washington, D.C., U.S.A.	72-405	38°51'N	77°02'W
Abastumani, U.S.S.R.	37-506	41°43'N	42°50'E
(Tbilisi, U.S.S.R.)	37-549	41°41'N	44°57'E
Rome, Italy	16-239	41°48'N	12°36'E
Vladivostok, U.S.S.R.	31-960	43°07'N	131°54'E
Alma Ata, U.S.S.R.	36-870	43°15'N	76°56'E
Green Bay, Wisconsin, U.S.A.	72-645	44°29'N	88°08'W
Bismarck, N. Dakota, U.S.A.	72-764	46°46'N	100°45'W
Arosa, Switzerland	---	46°47'N	09°41'E
(Milano, Italy)	16-080	45°28'N	09°17'E
Caribou, Maine, U.S.A.	72-712	46°50'N	68°00'W
Cambourne, England	03-808	50°13'N	05°19'W
Moosonee, Canada	72-836	51°16'N	80°39'W
Oxford, England	---	51°46'N	01°16'W
(Crawley, England)	03-774	51°05'N	00°13'W
Edmonton, Canada	72-879	53°34'N	113°31'W
Eskdalemuir, Scotland	03-162	55°19'N	03°12'W
(Leuchars, Scotland)	03-171	56°23'N	02°53'W
Aarhus, Denmark	06-070	56°18'N	10°37'E
(Copenhagen, Denmark)	06-180	55°38'N	12°40'E
Uppsala, Sweden	02-076	59°52'N	17°37'E
(Stockholm, Sweden)	02-077	59°21'N	17°57'E
Leningrad, U.S.S.R.	26-063	59°57'N	30°42'E
Lerwick, Scotland	03-005	60°08'N	01°11'W
Reykjavik, Iceland	04-030	64°08'N	21°54'W
(Keflavik, Iceland)	04-018	63°57'N	23°37'W
Resolute, Canada	72-924	74°43'N	94°59'W
Alert, Canada	74-082	82°30'N	62°20'W

TABLE 2

Horizontal Flux of Ozone by Transient Eddies
(cm of ozone at STP x m sec.$^{-1}$; no. of cases in parentheses)

Latitude belt or station	Jul-Sep 1957	Oct-Dec 1957	Jan-Mar 1958	Apr-Jun 1958	Jul-Sep 1958	Oct-Dec 1958
100 mb						
Marcus Island and Japan (4 stations)	-0.0664 (172)	-0.0050 (228)	-0.0532 (400)	+0.0040 (474)	-0.0418 (661)	-0.0555 (531)
35°-45° N (6 stations)	+0.0099 (161)	+0.0005 (184)	+0.0820 (232)	+0.0156 (419)	+0.0048 (398)	-0.0008 (332)
45°-55° N (7 stations)	+0.0518 (568)	+0.0510 (529)	+0.0725 (499)	+0.0043 (738)	+0.0339 (749)	+0.0461 (606)
55°-60° N (5 stations)	+0.0382 (310)	+0.0819 (261)	+0.0781 (380)	-0.0096 (548)	+0.0488 (546)	+0.0464 (345)
Keflavik 64° N	+0.0381 (140)	+0.0240 (125)	-0.1779 (136)	+0.0736 (167)	-0.0081 (166)	-0.1968 (105)
Resolute 75° N	+0.0647 (29)	+0.1022 (18)	-0.0421 (6)	+0.0383 (99)	+0.0442 (128)	+0.1111 (10)
Alert 82.5° N	+0.0064 (120)	+0.0049 (13)	-0.9798 (6)	+0.0183 (124)	+0.0220 (128)	+0.0752 (10)
50 mb						
Marcus Island and Japan (4 stations)	-0.0075 (148)	-0.0030 (211)	-0.0389 (324)	+0.0213 (373)	-0.0127 (536)	+0.0128 (453)
35°-45° N (6 stations)	+0.0039 (79)	+0.0282 (92)	+0.1175 (166)	+0.0083 (321)	+0.0049 (300)	-0.0210 (218)
45°-55° N (7 stations)	+0.0319 (409)	+0.0097 (388)	+0.1152 (372)	+0.0081 (568)	+0.0222 (592)	+0.0346 (518)
55°-60° N (5 stations)	+0.0188 (180)	+0.0683 (151)	+0.0783 (192)	+0.0103 (359)	+0.0319 (335)	+0.0970 (207)
Keflavik 64° N	+0.0169 (141)	+0.0572 (117)	-0.2680 (125)	+0.0390 (166)	+0.0030 (161)	-0.2420 (96)
Resolute 75° N	+0.0406 (25)	+0.1011 (13)	-0.0578 (4)	+0.0077 (87)	+0.0341 (110)	+0.2015 (14)
Alert 82.5° N	+0.0022 (113)	+0.0675 (5)	-0.4754 (7)	-0.0186 (126)	+0.0087 (121)	+0.0833 (9)

TABLE 3

Ozone Budget
(cm^3 of ozone at STP sec^{-3} when multiplied by 10^9)

	July-Sept. 1957	Jan-Mar. 1958	July-Sept. 1958
Transport of ozone across 50° N			
a) by transient eddies	+2.7	+9.9	+1.9
b) by standing eddies	+0.1	+3.7	-0.7
c) by mean meridional motions	-2.9	?	-2.9
Transport from content change north of 50° N	-5.2	+9.0	-5.2
Transport of ozone across 40° N			
a) by transient eddies	+0.3	+10.1	+0.4
b) by standing eddies	+0.1	+3.7	-0.7
c) by mean meridional motions	-4.3	?	-4.3
Transport from content change north of 40° N	-7.4	+11.6	-7.4
Tropospheric downward flux	~6		

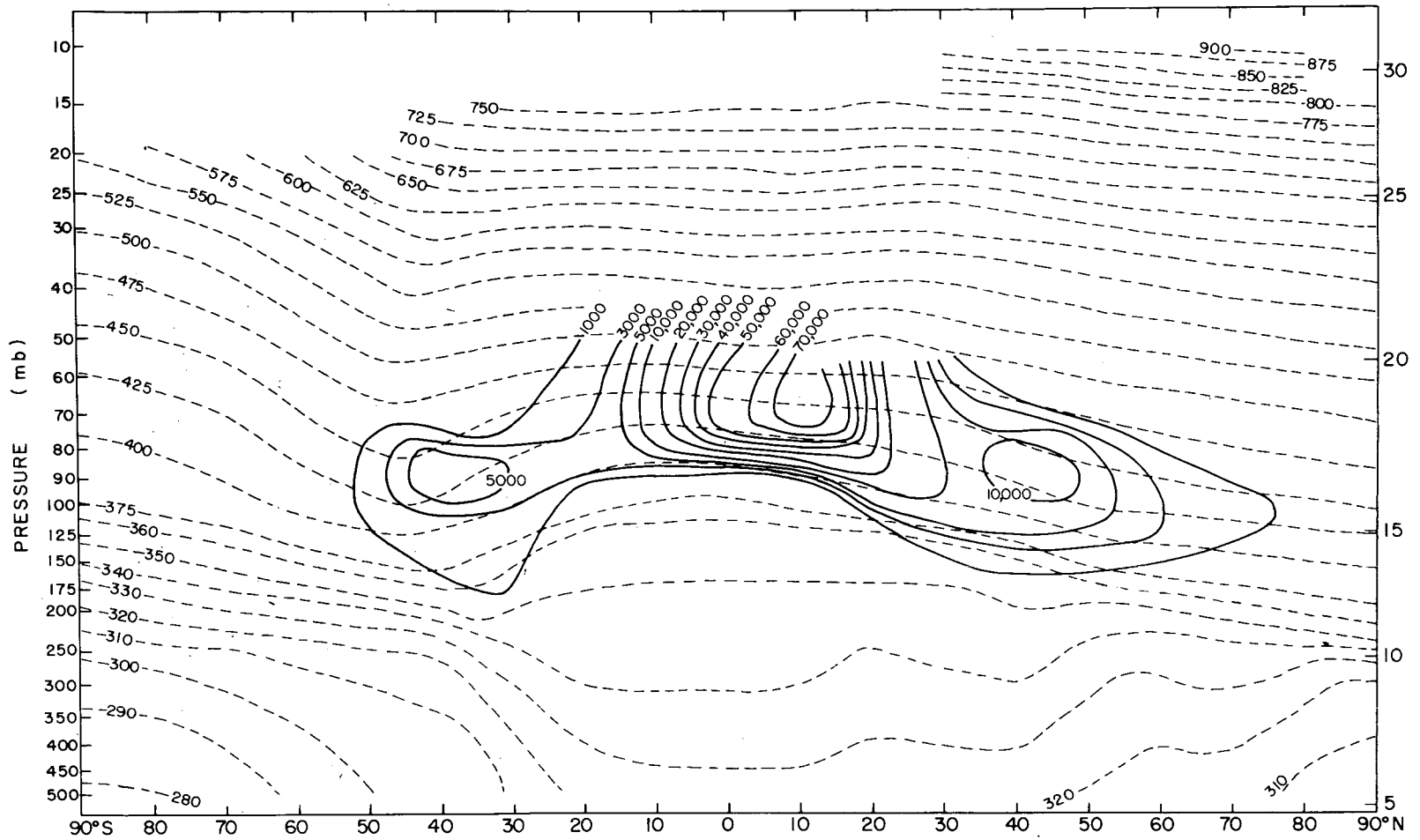

FIG. 1

ISENTROPES JULY, 1957

TUNGSTEN-185 (dpm/1000 SCF) SEPT.-OCT., 1958

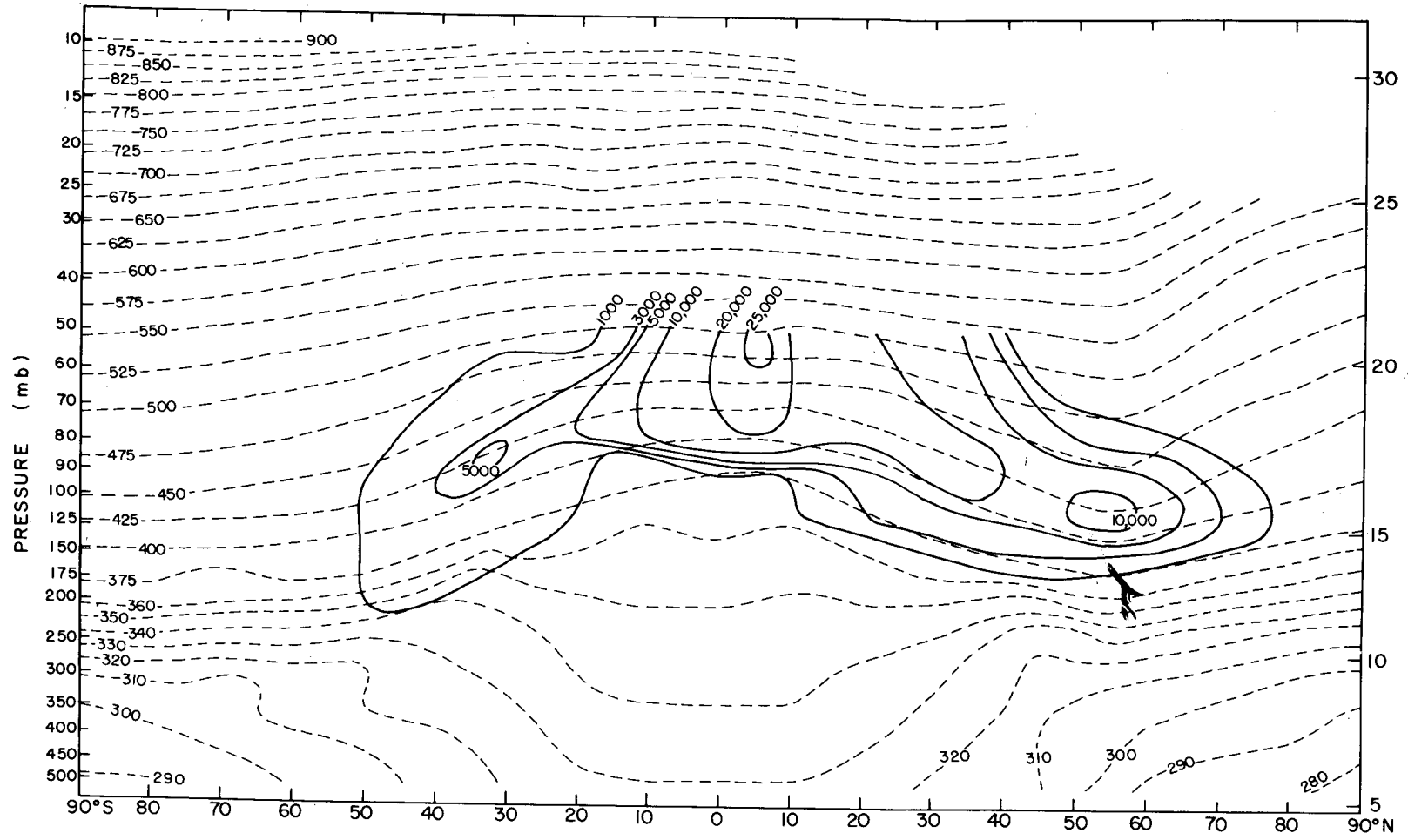

FIG. 2

ISENTROPES DEC., 1957

TUNGSTEN - 185 (dpm/1000 SCF) NOV. - DEC., 1958

GEOCHEMICAL STUDIES ON THE STRATOSPHERIC FALLOUT

P.K. Kuroda, H.L. Hodges, M.P. Menon, Tin Mo, Joe Nix, L.M. Fry and H.E. Moore*
Department of Chemistry
University of Arkansas
Fayettville, Arkansas

Introduction

This report gives a brief summary of the work being carried out in this laboratory on the radioactive fallout since the fall of 1958. The topics under investigation include: (a) nuclear fission in the early history of the earth, (b) spring peak of Sr^{90} fallout, (c) fallout from nuclear detonations of February and April 1960, (d) stratospheric residence time of Sr^{90}, and (e) stratospheric fallout of Ce^{144}.

Nuclear Fission in the Early History of the Earth

It has been pointed out by Kuroda(1), in 1958, that the Sr^{90} is produced in nature by the spontaneous fission of uranium and thorium and that the total amount of Sr^{90} produced by the spontaneous fission of U^{238} alone in the earth's crust is comparable to the amount of artificially produced Sr^{90} in the stratosphere.

After the mass spectrum of meteoritic xenon became known in 1960(2), Kuroda(3) pointed out that the differences between meteoritic and terrestrial xenon seemed to indicate an excess of the heavy isotopes of xenon (Xe^{131}, Xe^{132}, Xe^{134} and Xe^{136}) in the earth's atmosphere, and reported that the difference is much greater than that expected from the U^{238} spontaneous fission alone, but can be explained as due to the spontaneous fission of some of the extinct transuranium

*On leave from the Department of Chemistry, Arkansas State College, State College, Arkansas, during the summers of 1960 and 1961.

elements and/or the induced fission of U^{235} in the early history of the earth. It has been suggested that spontaneous fission of extinct Pu^{244} (7.6×10^7 years) might have produced the excess $Xe^{131-136}$ in the earth's atmosphere.

Kuroda(4) attempted to calculate the time interval between the cessation of the nucleosynthesis and formation of the earth, assuming that the extinct Pu^{244} was mainly responsible for the production of excess xenon isotopes in the earth's atmosphere and obtained a value of 5.2×10^7 years for the time interval.

Kuroda(5) has recently revised the calculation, and showed that the calculated time interval between the cessation of nucleosynthesis and formation of the earth based on the Pu^{244} model to be most likely approximately 10^8 years.

Another important source of contribution to the atmospheric inventory of xenon is the neutron-induced fission of U^{235} in nature. A number of reports have been published on this subject (6)(7)(8), but the exact contribution from this source can not be evaluated at the present time.

According to Kuroda(3), at least 10% of the total amount of Xe^{136} in the earth's atmosphere is "fissiogenic". It is interesting to note that the contribution from the recent artificial nuclear explosions is estimated to be less than only 10^{-6} of the contributions from the natural fission processes.

Spring Peak of Sr^{90} Fallout

To explain the vertical distributions of water vapor and ozone in the atmosphere, Brewer(9), in 1949, and Dobson(10), in 1956, proposed a model of global movement of the air masses. According to this model, there is a cold pool of air in the stratosphere over the winter pole during the late winter months, and it carries ozone-rich air to the lower levels in early spring. It was further suggested by Dobson that, if there is such a slow sinking of air in the middle latitudes from the stratosphere to the troposphere, it must be balanced elsewhere by a reserve current from the troposphere to the stratosphere. This upward flow of air is expected to occur in tropical regions.

Experimental results obtained in this laboratory seemed to support the Brewer-Dobson model of global circulation of air masses quite well. A pronounced peak of fallout rate occurred at Fayetteville, Arkansas, during the spring months of 1959(11). We concluded that

the spring peak was primarily due to an increase in the rate of material transfer from the stratosphere to the troposphere, caused by the sinking of every cold air masses which formed above the winter pole during the late winter months, and we predicted, in 1959, that there would be another peak in the Sr^{90} fallout rate during the spring months of 1960, regardless of whether or not there was a thermonuclear bomb explosion during the fall months of 1959.

The 1960 spring peak, clearly shown in Fig. 2, demonstrated that the seasonal and global movements of stratospheric air masses, such as described by Brewer and Dobson, play an extremely important role in causing the spring peaks of the Sr^{90} fallout.

Our data seem to be explained quite well by the assumption that whereas the transfer from the stratosphere occurred at a maximum rate in the spring months and at a minimum rate in the fall months, the overall rate of the Sr^{90} transfer from the stratosphere during the period 1959-1960 was roughly equivalent to an "apparent" stratospheric mean storage time of approximately 1 year or even less.

Fallout from Nuclear Detonations of February and April 1960

A sharp increase in the Sr^{89}/Sr^{90} ratio in rain was observed at Fayetteville, Arkansas, after the French nuclear detonations of February and April 1960, as shown in Fig. 1(12).

We have interpreted the strontium isotope ratio data and concluded that (a) the Sr^{90} inventory of the Northern Hemisphere has increased due to the French nuclear explosions by approximately 0.1 to 0.2% and (b) this is due to the fact that there is an upward flow of air in the tropical region and hence a material transfer from the troposphere to the stratosphere is expected to occur, according to the global circulation model of air masses, proposed by Brewer(9) and by Dobson(10).

It has been generally accepted that relatively low yield nuclear bombs such as those detonated at Reggan in the Sahara Desert on February 13 and on April 1, 1960, inject their debris solely into the troposphere, whereas debris from high yield thermonuclear bombs enter the stratosphere and cause the worldwide stratospheric fallout.

Although the French nuclear detonations caused seemingly small transient increases in fallout and the fresh debris most likely added no more than a few tenths percent to the total worldwide fallout of long-lived fission products, the radionuclide injection into the atmosphere by the French nuclear detonations was of unique scientific interest in that the nuclear explosions occurred in the

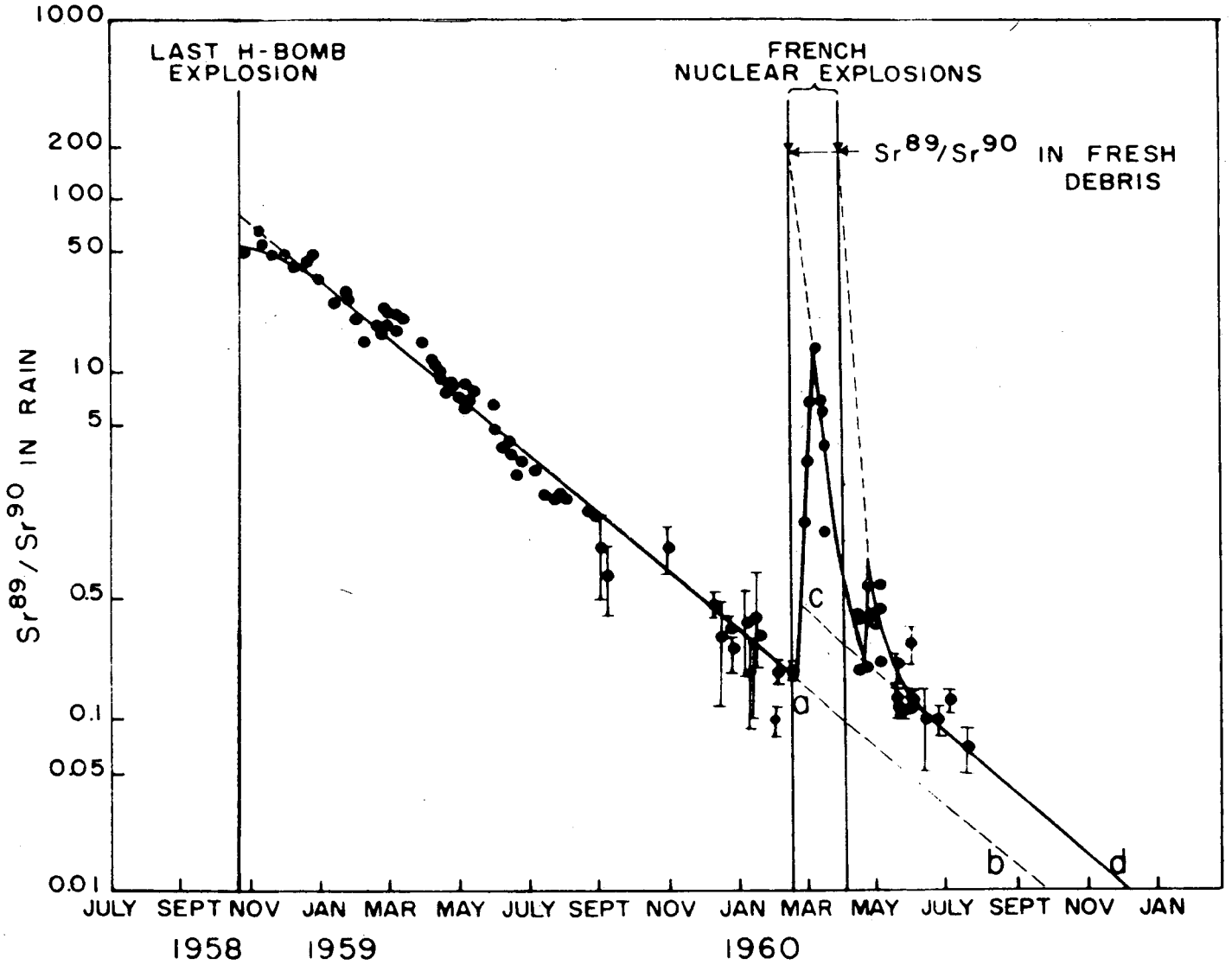

Figure 1. Variation of the Sr^{89}/Sr^{90} ratio in individual samples of rain and snow collected at Fayetteville, Arkansas, since November 1958

tropical region after a longer nuclear test suspension period.

Stratospheric Residence Time of Sr^{90}

Widely different values have been reported in the past for the stratospheric residence time of Sr^{90}. Libby (13) first pointed out the long hold-up of Sr^{90} in the stratosphere and estimated the mean stratospheric residence time to be 5 to 10 years. Machta and List (14) estimated the mean removal rate to be about 20% per year, but later pointed out the possibility that the actual rate might be much greater.

Kulp et al. (15) proposed a value of 3 years for the residence time, and Storebø (16) reported that the residence time in the stratosphere should not be much more than 1 year, while Feely (17), in 1960, estimated the residence half-time to be less than 1 year, equivalent to a mean residence time of less than 18 months.

A number of investigators (18)(19), on the other hand, have recently pointed out that the concept of a well-mixed stratosphere and a mean stratospheric residence time appear largely inapplicable to the interpretation of stratospheric fallout. Martell and Drevinsky (18)(19) proposed three stratospheric residence times instead of one: (a) a few months or more for Soviet test debris in the polar stratosphere; (b) 1 to 3 years for debris in the lower equatorial stratosphere; (c) 5 to 10 years for the debris at higher levels near the equator. Libby (20 has also expressed a similar view).

All these estimates were made on the basis of the experimental data obtained during the period when the nuclear test explosions were frequently conducted at various localities of the world. The magnitude and the types of weapons exploded were usually unannounced, and a certain amount of guess work was involved in the calculations as to the amounts of Sr^{90} injected by these bombs at different times.

We have attempted to calculate the stratospheric mean residence time from the Sr^{90} concentration data in rain obtained in this laboratory during the nearly three year test "suspension" period.

Monthly average Sr^{90} concentrations in rain ($\bar{C}$) were calculated from the equation,

$$\bar{C} = F/R \quad \ldots\ldots\ldots\ldots\ldots (1)$$

where F is the total amount of Sr^{90} (in $\mu\mu c/m^2$) transported by rain

TABLE 1

Monthly Average Sr^{90} Concentrations in Rain at Fayetteville, Arkansas ($\mu\mu c/l$)

Month	$\bar{C}$	Month	$\bar{C}$
November 1958	2.90	April 1960	3.23
December 1958	6.97	May 1960	1.92
January 1959	11.4	June 1960	3.35
February 1959	10.7	July 1960	0.75
March 1959	13.8	August 1960	1.13
April 1959	24.1	September 1960	0.84
May 1959	18.7	October 1960	0.51
June 1959	5.63	November 1960	0.30
July 1959	4.82	December 1960	0.44
August 1959	2.02	January 1961	1.43
September 1959	0.80	February 1961	2.11
October 1959	3.35	March 1961	2.44
November 1959	0.3	April 1961	3.51
December 1959	2.36	May 1961	1.86
January 1960	1.04	June 1961	1.71
February 1960	3.79	July 1961	0.86
March 1960	2.98	August 1961	0.65

during a period of a month and $\sum R$ is the total rainfall (in mm) during the same period. The values of $\bar{C}$ are shown in Table 1.

The data show that there is a marked seasonal variation of the Sr^{90} concentration in rain which follows a cyclic pattern with a maximum in the spring and a minimum in the fall.

The values of monthly average concentration of Sr^{90} in rain are not too accurate since the amounts and frequencies of rainfalls vary from month to month. For example, the value of $\bar{C}$ for the month of November 1959 is based on the measurement of a single rainfall. An exceptionally dry month may often be followed by an unusually wet month. For this reason, it has been decided to calculate the bi-monthly average concentrations by simple taking the arithemtic average of the values of $\bar{C}$ for two consecutive months.

The values of bi-monthly average Sr^{90} concentrations for the first year ($\bar{C}_1$), second year ($\bar{C}_2$) and third year($\bar{C}_3$) are shown in Table 2.

These values are plotted in Fig. 2. Values observed by Kuroda (1) in 1958 at Lemont, Illinois, are shown for comparison.

The percentage increase of the stratospheric Sr^{90} inventory in the Northern Hemisphere due to the French nuclear detonations of February and April 1960 was calculated to be approximately 0.1 to 0.2 % (12). Thus, we may neglect the contributions from a number of small French nuclear detonations, which occurred during the nuclear test "suspension" period, in the following calculation.

Contributions from the tropospheric fallout of the French tests and the fall 1958 Soviet tests appear to have been less than a few percent and hence should not affect the following calculation considerably.

Let us now compare the by-monthly Sr^{90} concentration during a two-month period with that during the same two-month period of the following year.

The average Sr^{90} concentrations in rain will certainly depend upon various factors, such as the types, total amounts and frequencies of rainfalls. However, let us by way of experiment write that

$$\bar{C}_{n+1} / \bar{C}_n = S_{n+1}/S_n \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (2),$$

TABLE 2

Bi-Monthly Average Sr^{90} Concentrations in Rain at Fayetteville, Arkansas
($\mu\mu c/l$)

Period	$\bar{C}_1$	Period	$\bar{C}_2$	Period	$\bar{C}_3$
November and December 1958	4.95	November and December 1959	1.33	November and December 1960	0.37
January and February 1959	11.1	January and February 1960	2.42	January and February 1961	1.77
March and April 1959	19.0	March and April 1960	3.11	March and April 1961	2.98
May and June 1959	12.2	May and June 1960	2.64	May and June 1961	1.79
July and August 1959	3.42	July and August 1960	0.94	July and August 1961	0.75
September and October 1959	2.08	September and October 1960	0.58		

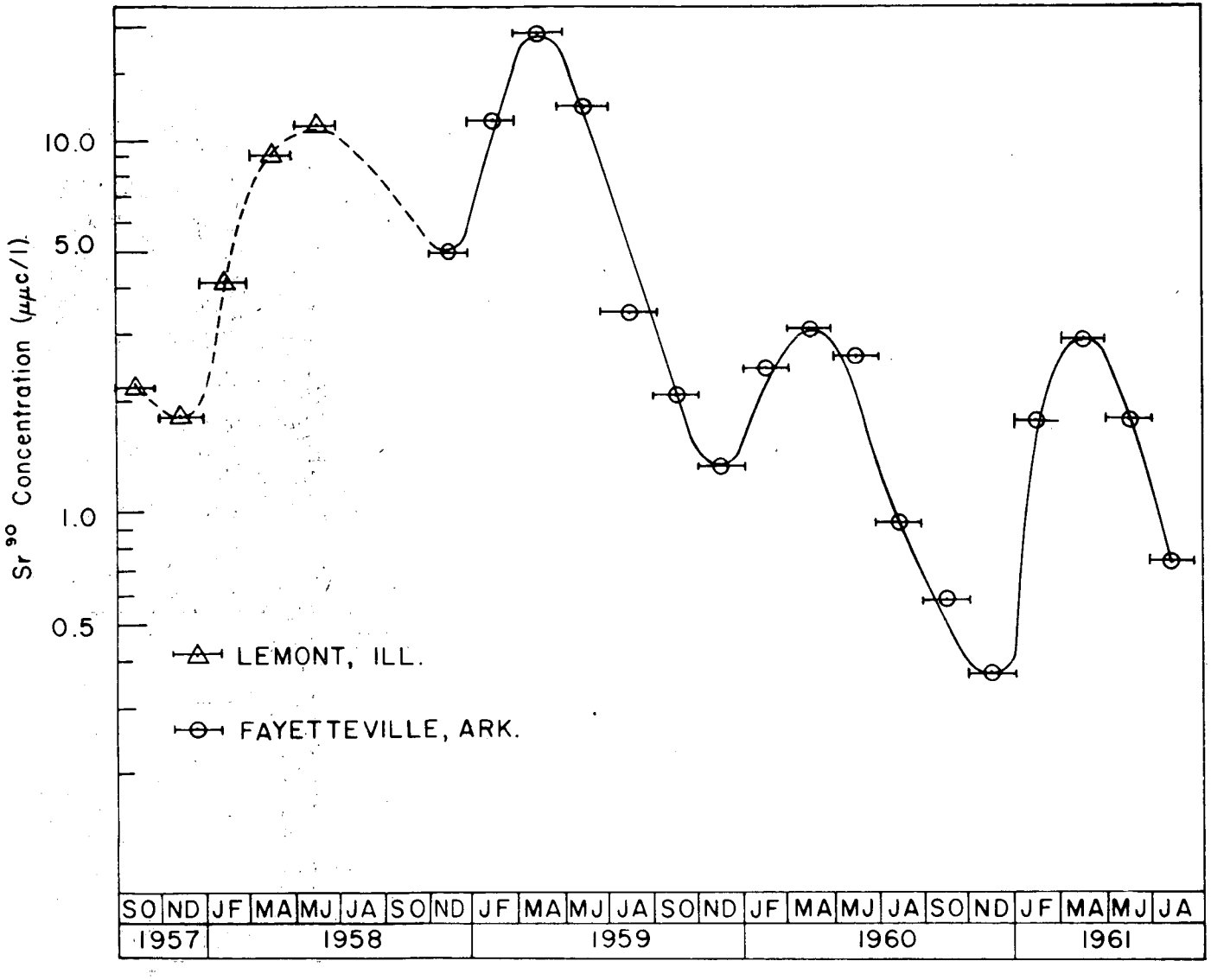

Figure 2. Bi-monthly Average Sr^{90} Concentrations in Rain

where S_n is the Sr^{90} inventory in the stratosphere during a two-month period in the nth year, S_{n+1} is that in the corresponding period in the (n+1)th year, and n=1 or 2.

The value of S_{n+1} will ordinarily be smaller than the value of S_n, because Sr^{90} in the stratosphere will be removed by the fallout and by the radioactive decay. Hence, we may write that

$$S_{n+1} = S_n \cdot e^{-(\bar{a} + \lambda_{90})} \quad \ldots\ldots\ldots\ldots (3)$$

where λ_{90} is the decay constant of Sr^{90} and $\bar{a}$ is a value which depends upon the annual average rate of stratospheric fallout.

It has to be noted that nothing has been said here about whether or not the stratosphere contains radioactivity which is uniformly mixed. In other words, we are by no means trying to give here what might ordinarily be described as an explanation for the mechanism of radioactive fallout.

The value of $\bar{a}$ may or may not be a constant depending upon the mechanism of radioactive fallout and also the "homogeneity" of the distribution of Sr^{90} in the stratosphere. It is entirely possible that even a negative value of $\bar{a}$ might be obtained if the stratosphere is not well mixed. Our next step is then to calculate the values of $\bar{a}$ from the values of $\bar{C}_1$, $\bar{C}_2$ and $\bar{C}_3$.

From equations (2) and (3), we have

$$\bar{C}_{n+1} = \bar{C}_n \cdot e^{-(\bar{a} + \lambda_{90})} \quad \ldots\ldots\ldots\ldots (4).$$

The values of $\bar{a}$ are calculated from the data given in Table 2 and are shown in Table 3.

It is interesting to note that the value of $\bar{a}$ remained fairly constant for almost two years, and then a sudden change to a new set of values took place after January 1961. The constancy of the value of $\bar{a}$ was as good as could be expected, however, during the first two-year period considering the many uncertainties involved in the treatment of the experimental data. For this reason, we have recently proposed (21) to define $(1/\bar{a})$ as the mean residence time and $\ln(2/\bar{a})$ as the residence half-time of Sr^{90} in the stratosphere. A value of 0.7 ± 0.1 years was obtained from the data available at that time for the mean residence time of Sr^{90}.

TABLE 3

Mean Residence Time and Residence Half-time of Sr^{90} in the Stratosphere (years)

Period	$\bar{a}$	Mean residence time, $1/\bar{a}$	Residence half-time
November and December 1958-1959	1.28	0.78	0.54
January and February 1959-1960	1.49	0.67	0.46
March and April 1959-1960	1.78	0.56	0.39
May and June 1959-1960	1.49	0.67	0.46
July and August 1959-1960	1.26	0.79	0.55
September and October 1959-1960	1.25	0.80	0.55
November and December 1959-1960	1.30	0.78	0.53
January and February 1960-1961	0.29	3.4	2.4
March and April 1960-1961	0.02	~50	~35
May and June 1960-1961	0.37	2.8	1.9
July and August 1960-1961	0.22	4.6	3.2

Excluding the March-April 1960-1961 value for $1/\bar{a}$, we now have an additional set of values roughly corresponding to 3.6 ± 0.9 years, which is quite in line with the values often mentioned by early investigators.

Perhaps a few words may have to be added here concerning the anomalous value from the March and April 1960-1961 data. We note in Table 3 that the value of $1/\bar{a}$ calculated from the March and April 1959-1960 data is also somewhat out of line and definitely lower than the average value of $1/\bar{a}$ for the first two-year period.

Suppose the value of $\bar{C} = 3.11$ μμc/l for March and April 1960 was too low for some reason. If it should have been 4 μμc/l, instead, then we have the following new values which can be beautifully incorporated in Table 3 as substitutes.

	$\bar{a}$	$1/\bar{a}$(year)
March and April 1959-1960	1.53	0.65
March and April 1960-1961	0.27	3.7

Such an error in obtaining the value of $\bar{C}$ can arise from various sources. To make sure that it is not due to analytical errors, we are now repeating the measurements of Sr^{90} in some of the March and April 1960 and 1961 rain samples. It is interesting to calculate an "overall apparent" mean residence time during the nuclear test suspension period by simply considering the ratio $\bar{C}_{n+2}$ and $\bar{C}_n$, i.e.,

$$\bar{C}_{n+2} = \bar{C}_n \cdot e^{-(\bar{a} + \lambda 90)(n + 2-n)} \quad \ldots . \quad (5).$$

The values of $\bar{a}$ are calculated from $\bar{C}_1$ and $\bar{C}_3$, and are shown in Table 4.

The constancy of the values of $\bar{a}$ thus obtained is again as good as could be expected. A value of 1.1 ± 0.3 years is thus obtained for the over-all apparent mean residence time. This value is similar to the estimates made earlier by Kuroda, Hodges and Moore (12), Storobø (16) and Feely (17).

TABLE 4

Over-all Apparent Mean Residence Time and Residence Half-time of Sr^{90} in the Stratosphere during the Nuclear Test Suspension Period 1958-1961 (years)

Period	$\bar{a}$	Mean residence time, $1/\bar{a}$	Residence half-time
November and December 1958-1960	1.27	0.79	0.55
January and February 1959-1961	0.89	1.12	0.78
March and April 1959-1961	0.90	1.11	0.77
May and June 1959-1961	0.93	1.08	0.75
July and August 1959-1961	0.73	1.37	0.95

Stratospheric Fallout of Ce^{144}

In connection with the observations on the Sr^{90} spring peaks, it was felt worthwhile to investigate the fallout pattern of Ce^{144} in some detail and we have recently undertaken the measurements of the Ce^{144} concentrations in individual samples of rain.

A new radiochemical procedure for cerium, which has recently been developed in this laboratory (22), was used for the determination of Ce^{144} in rain water. The new method depends entirely on the oxidation-reduction and liquid-liquid extraction cycle, and was originally intended for the fission products from the U^{238} spontaneous fission. Values of $7.0 \pm 1.4\%$ were secured by this method for the yields of Ce^{143} and Ce^{144}, respectively, in good agreement with the values obtained by Russell for the yields of Pr^{143} and Ce^{144} from the spontaneous fission of U^{238} (23).

Yields of cerium and strontium isotopes from fast neutron-induced fission of uranium were determined by irradiating a sample of depleted uranium oxide (99.989% U^{238} and 0.011% U^{235}) with $14.5 + 0.4$ Mev neutrons produced by the T(d,n) He^4 reactions in the University of Arkansas 400 KV Cockcroft-Walton positive ion accelerator (24)(25) and the following results were obtained: Ce^{144}, $5.2 \pm 0.4\%$; Sr^{90}, $3.4 \pm 0.3\%$. These results are in agreement with the values of fission yields for fast neutron-induced fission of U^{238} reported by Katcoff (26); Ce^{144}, 4.9%; Sr^{90}, 3.2%.

Although our value for the Ce^{144} yield is considerably higher than the value of $2.68 \pm 0.16\%$ reported by Cuninghame (27), and hence the experiments may have to be repeated, it is of some interest to use our data to calculate the Ce^{144}/Sr^{90} production ratio in the instantaneous fission of U^{238} caused by fast neutrons;

$$\frac{5.2}{3.4} \times \frac{28 \times 365}{285} = 54.8 \text{ (dpm/dpm)}$$

The monthly average Ce^{144} concentrations in rain were computed from the formula

$$\bar{C} = \Sigma F / \Sigma R,$$

where ΣF is the total amount of Ce^{144} (in $\mu\mu c/m^2$) transported by rain during a one-month period and ΣR is the total rainfall (in mm) during the same period.

The existence of a pronounced spring peak in the Ce^{144} fallout rate is obvious in Fig. 3. Considering the decay of Ce^{144} the 1961

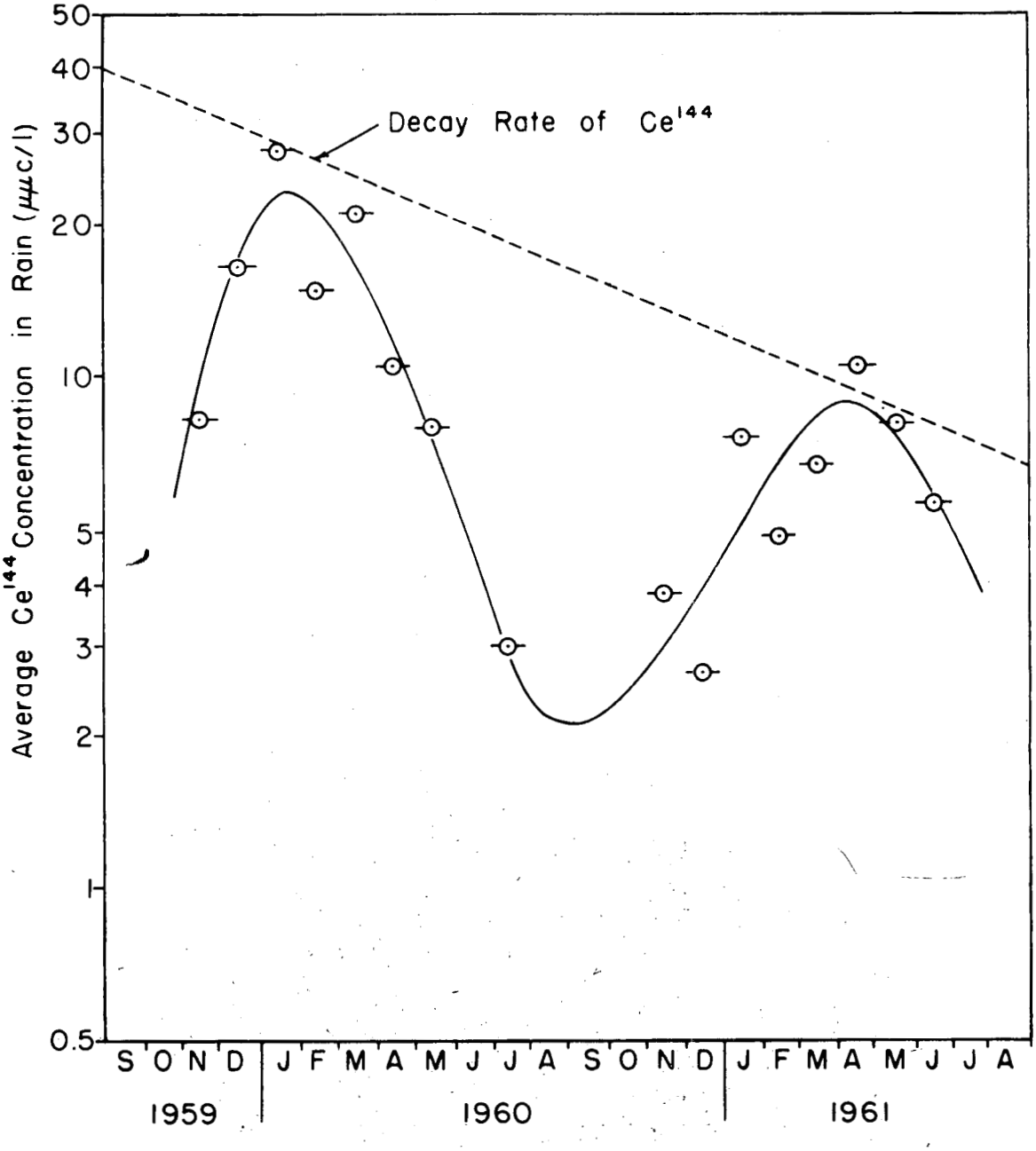

Figure 3. Seasonal Variation of the Ce^{144} Concentration in Rain at Fayetteville, Arkansas

spring peak was just as high as the 1960 peak. Thus, it looks as if the depletion of the Ce^{144} inventory by the fallout did not take place. A possible explanation for this may be that new debris may have been entering the lower stratosphere during this period at a rate similar to that of removal of Ce^{144} by the fallout process.

Fig. 4 shows the seasonal variation of the average monthly Ce^{144}/Sr^{90} ratio follows a cyclic pattern with a minimum in the spring and a maximum in the fall. It thus appears as if we are dealing with two stratospheric air masses; one containing Ce^{144} -rich and the other Ce^{144}-depleted debris. The cause of the spring minimum of the Ce^{144}/Sr^{90} ratio may be explained according to the Brewer-Dobson model (9)(10), at least qualitatively, as due to the presence of Ce^{144}-depleted debris at a very high altitude. As a cold air mass produced in the high stratosphere in the early spring slowly descends to lower levels to cause the spring peak of fallout, the Ce^{144}-depleted debris may show up in the lower stratosphere since the bulk of debris from the fall 1958 Soviet test series must have been largely removed from the lower stratosphere during the latter half of the nuclear test suspension period.

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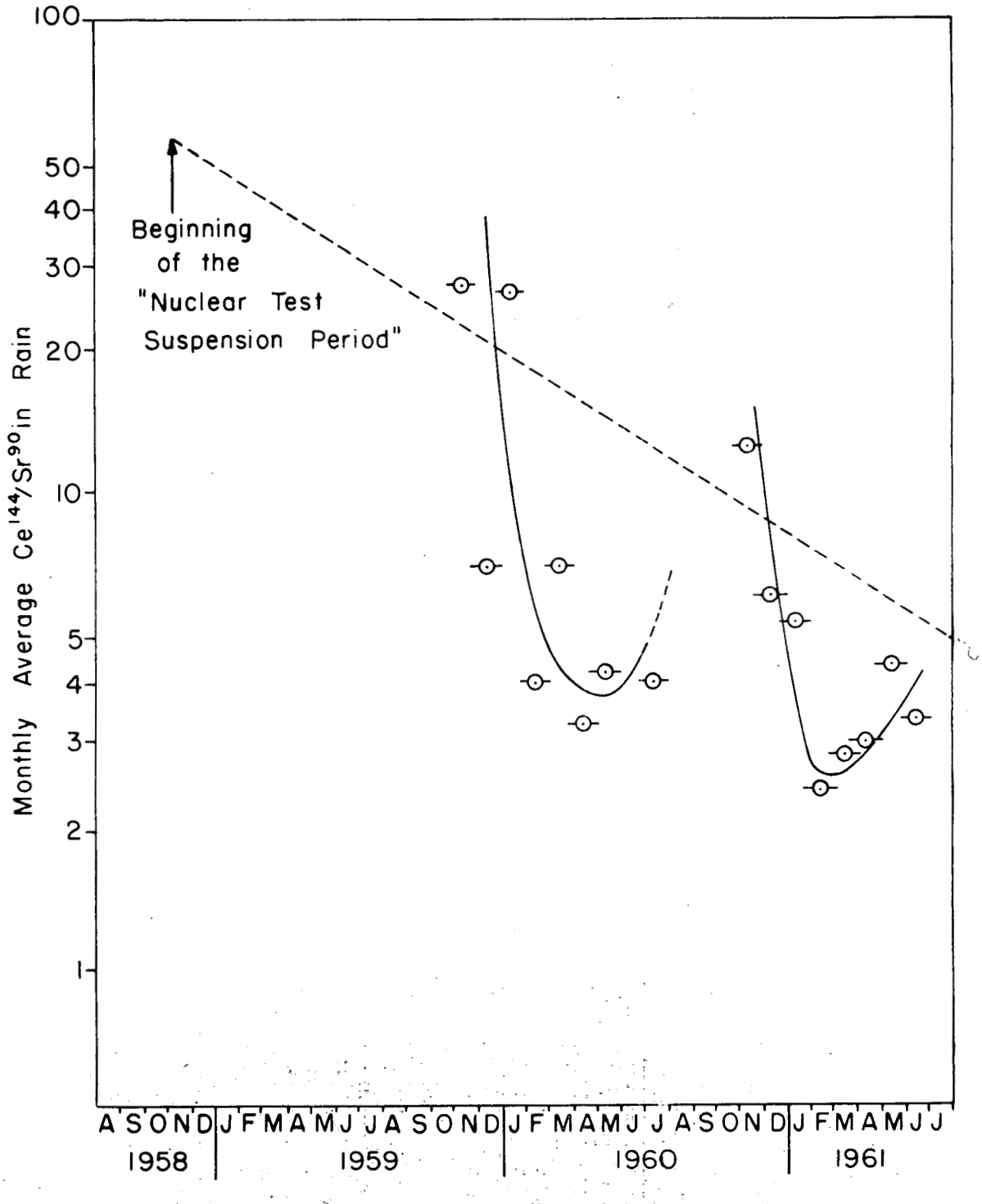

Figure 4. Variation of the Ce^{144}/Sr^{90} Ratio in Rain at Fayetteville, Arkansas. Dotted line I shows the calculated change of the Ce^{144}/Sr^{90} ratio in the stratosphere with time, which must be observed if an imaginary fresh debris with the initial Ce^{144}/Sr^{90} ratio = 54.8 was injected into the stratosphere in the fall of 1958, under the assumptions that (a) the stratosphere is uniformly mixed and (b) it was free of older debris.

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METEOROLOGICAL INFLUENCES ON SR^{90} FALLOUT CONCENTRATIONS IN PRECIPITATION
Part 1. LARGE--SCALE UPLIFT

Paul Kruger,** Leonard P. Salter,***and Charles L. Hosler ****
Nuclear Science and Engineering Corporation
Pittsburgh, Pennsylvania

Introduction and Summary

A program* is underway to study the dependence of fallout concentration in ground-level precipitation upon the meteorological conditions involved. Three types of precipitation-producing systems are being examined under this program: (1) large scale uplift, (2) convective storms, and (3) orographic systems. Precipitation is collected continuously during storms resulting from these systems and analyzed for Sr^{90} concentration. Meteorological data are collected for each storm studied. Correlations of the fallout concentration in the precipitation with the meteorological data are made as a function of time.

A preliminary investigation (1) has been completed for the first of the three weather systems. Analysis of three winter rainstorms resulting from large-scale uplift of air in which the generating level and the precipitation mechanism were essentially constant shows that the ground-level Sr^{90} concentration in the precipitation is determined by the descent

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** Hazleton-Nuclear Science Corporation, Palo Alto, California. Senior investigator under subcontract.
*** Present address: Health and Safety Laboratory, U. S. AEC, New York.
**** State College, Pennsylvania. Consultant to Nuclear Science and Engineering Corporation.

experience of the precipitation in falling from the generating cloud to the ground. The descent experience can be expressed in terms of the height of the ceiling, which is an index of the humidity profile and depth of the lower layers of air. These parameters affect the rate of evaporation or growth of the precipitation elements, and thus, the final concentration of Sr^{90} reaching the ground.

Work is presently in progress to study the influence of convective showers on the Sr^{90} fallout concentration. In contrast to the large-scale uplift of air in frontal systems, convective showers present a rapidly changing generating level with vertical air motions of the same magnitude as the horizontal motions. Further, since the drop sizes of rain falling from convective showers are much larger than those from stable air, the effect of evaporation during the descent will not be as important.

Study of orographic systems will be made this winter during the rainy season in California to examine the downwind change in ground-level Sr^{90} concentrations resulting from steady-state stable precipitating cloud systems. An investigation may also be made of the hypothesis of Storebo(2) that selective adherence of different nuclides to different size particles, acting as nucleating agents within the clouds, may result in measurable differences of such radionuclide ratios in the rain collected at the low and high locations along the orographic trajectory.

This paper discusses the overall program and the results obtained for the first of the three weather systems, the large-scale uplift of air associated with frontal systems. Preliminary data are also given for the convective storms studied to date.

Techiques and Data Sought

The study of the meterological influences on fallout concentrations in precipitation is performed by correlation of Sr^{90} concentration with the measured and computed meteorological data available for each storm.

Rainfall collection systems of several types are used for obtaining the precipitation samples. The collection system for the large-scale uplift studies has been described previously.(3,4) The samples are analyzed by standard radiochemical procedures for low levels of Sr^{90} and other radioisotopes. The average spread between duplicate assays of Sr^{90} has been shown to be about 10 to 15%(3). This precision is considered adequate for the current program since there are greater variabilities in the meteorological data.

Rainfall rate data are collected at the collection site; and the average precipitation rate for each sample interval is calculated from the sample volume, the collection area, and the time interval of precipitation.

Other meteorological information is collected at the collection site, and a meteorological analysis is performed of these and data obtained

from the U. S. Weather Bureau and the Federal Aviation Agency. Future studies will include radar observations made at the collection sites, where possible.

For the completed studies of the large-scale uplift systems, time sections for the hourly weather situation were produced. Regular 6- and 12-hour surface-and upper-air analyses were obtained from the facsimile circuit of the U.S. Weather Bureau Analysis Center. In addition, hourly 1-mb surface analyses were made covering all periods of data collection. From these analyses trajectories were computed to determine the precipitation history of air near the surface and at the generating level. Time cross-sections of the vertical distribution of cloudiness over Pittsburgh, Pennsylvania, were made. The height of the ceiling and the thickness of the low clouds were also tabulated during the data collection periods. The types of data obtained for each storm are listed in Table 1. Fig. 1 and Tables 2 and 3 show as examples the data obtained(1) for Storm No. 1.

TABLE 1

Data obtained for storms from
large-scale uplift of stable air

1.	Description of storm system
2.	Surface and 500-mb charts near start of precipitation
3.	Sample collection periods
4.	Type and periods of precipitation
5.	Quantity of precipitation
6.	Precipitation rate
7.	Amount of Sr^{90} in sample
8.	Sr^{90} concentration
9.	Hourly ceiling measurements
10.	Hourly previous precipitation experience at low levels
11.	Hourly previous precipitation experience at the generating level

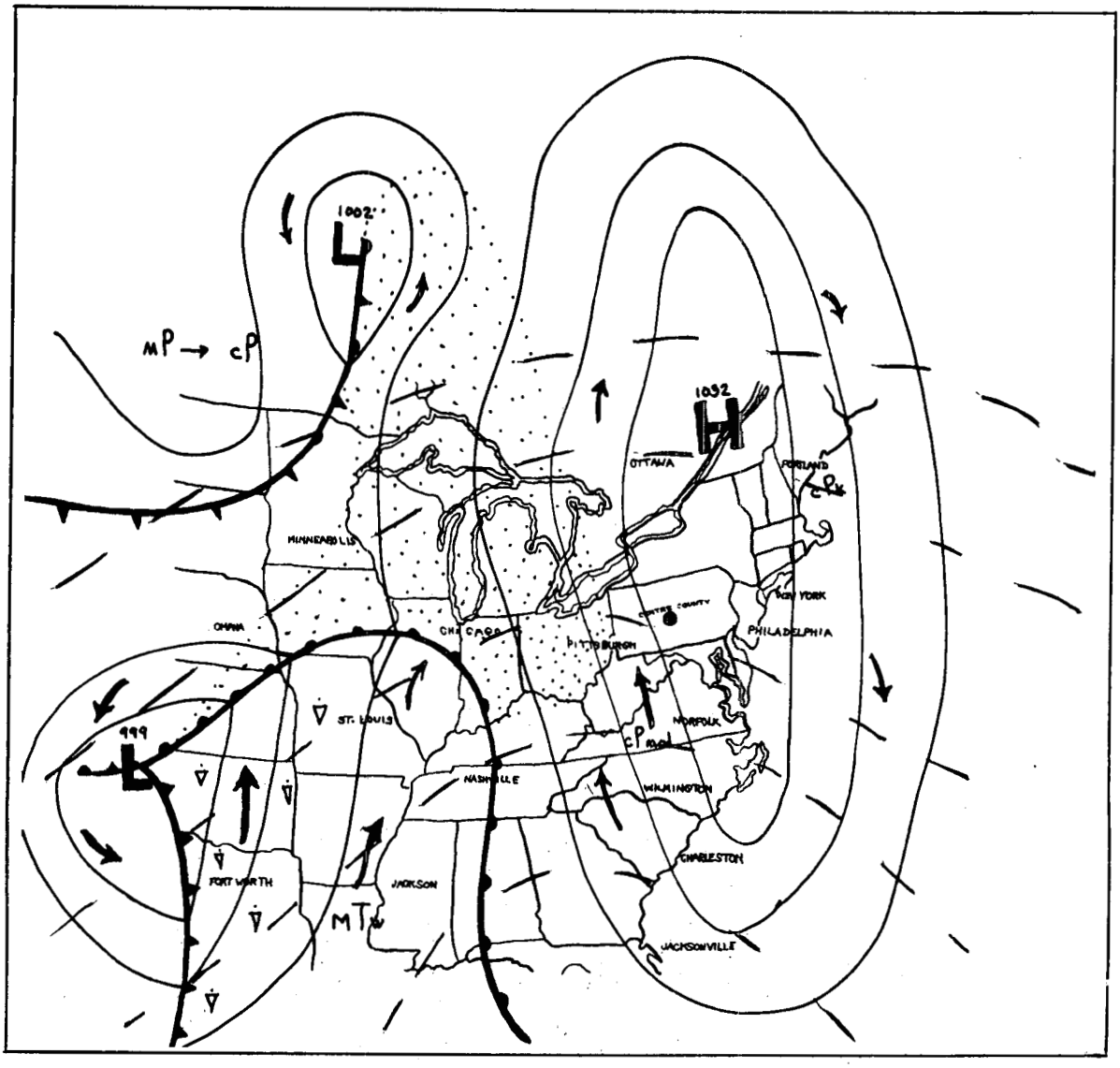

Fig. 1. Surface Map 7 am Jan. 12, 1960
(500 mb flow indicated by dashed lines)

TABLE 2

Precipitation Collection and Sr^{90} Concentration Data - Storm No. 1 - 12 January 1960

Collection Period From Day	Time	To Day	Time	t* (hrs.)	Precipitation (rain) (liters)	(inches)	Prec. Rate (inches/hour) Ave.	Max.	Min.	Sr^{90} (dpm/sample)	Sr^{90} (dmp/liter)
10 Jan	1500	12 Jan	0925	1.4	0.15	0.008	0.006	0.009	0	3.3±0.6	22±4
10 Jan	1500	12 Jan	0925	1.4	0.15	0.008	0.006	0.009	0	3.6±0.8	24±6
12 Jan	0925	12 Jan	1201	2.60	0.85	0.047	0.018	0.059	0.007	5.0±0.9	5.9+1.1
12 Jan	0925	12 Jan	1211	2.77	1.09	0.061	0.022	0.083	0.007	9.4±1.0	8.6±0.9
12 Jan	1201	12 Jan	1330	1.48	3.7	0.21	0.142	0.26	0.046	5.6±0.8	1.5±0.2
12 Jan	1211	12 Jan	1336	1.42	4.0	0.22	0.155	0.26	0.075	8.7±1.7	2.2±0.4
12 Jan	1330	12 Jan	1418	0.80	6.6	0.37	0.462	0.87	0.20	2.9±0.8	0.44±0.12
12 Jan	1336	12 Jan	1423	0.78	6.8	0.38	0.487	0.87	0.20	1.4±0.6	0.21±0.09
12 Jan	1423	12 Jan	1509	0.77	2.0	0.114	0.135	0.22	0.083	5.5±1.5	2.7±0.7
12 Jan	1418	12 Jan	1634	2.77	3.7	0.21	0.093	0.25	0	9.7±0.9	2.6±0.2
12 Jan	1509	12 Jan	1509	1.60	1.4	0.077	0.048	0.14	0	3.1±1.3	2.2±0.9

Collections for the convective shower studies will be made at State College, Pennsylvania. In addition to the meteorological data taken during the large-scale uplift studies radar observations are being made at this site to locate the height of the generating level and the horizontal extent of the storms during the precipitation periods.

For the orographic studies, initial collections will be made this winter with the onset of the rainy season. Arrangements have been made to obtain meteorological coverage of the orographic storms which occur on the lower coastal ranges near Santa Barbara, California. Collections are scheduled to be made at three locations along the trajectory of the system: at the coast near sea level, at the top of the Santa Ynez Mountains (4000-feet elevation) and at a site midway up the mountain. Special plastic collectors with an area of 400 square feet will be used to obtain the precipitation samples at these three sites. Radar coverage of these storms also is desired.

TABLE 3

Ceiling and previous precipitation experience
Storm No. 1 - 12 January 1960

Time	Ceiling (feet)	Previous precipitation experience (hours)	
		Surface	Generating level
12 Jan 60			
0700	>20,000	0	6
0800	>20,000	1	6
0900	3,600	2	7
1000	3,000	3	7
1100	6,000	4	7
1200	4,000	5	7
1300	2,500	6	10
1400	200	7	10
1500	400	8	8
1600	500	4	6
1700	2,000	3	3

Large-Scale Uplift of Air

In general, the fallout radioactivity concentration in precipitation which reaches the ground is likely to be dependent upon the parameters[1] listed in Table 4.

For storms resulting from large-scale uplift of air, the first two variables may be considered to be constant. For the storms studied in this program[1], the precipitation is formed in the ice phase at approximately the same altitudes. Further, by limiting observations to precipitation involving one air mass, and since short period changes in water content within air masses are small, the fallout and water concentrations would not be expected to change significantly. Therefore, attention was focused upon the two remaining variables.

The possible dependence of the Sr^{90} concentration upon the previous precipitation history at the generating and lower levels and the descent experience have been described.[1] For the three storms resulting from large-scale uplift of air, the Sr^{90} concentration in the collection samples was compared with the hours of previous precipitation experience at the generating level above the collection site, as computed from the synoptic data, for studying the fifth variable listed in Table 4; and

TABLE 4

Parameters likely to influence fallout concentrations in precipitation

1. The height of the precipitation generating level
2. The precipitation generation and growth mechanisms in the cloud
3. The amount of radioactivity initially in the air masses participating in the precipitation process
4. The specific humidity at the generating level
5. The previous precipitation experience of the air at the generating level
6. The descent experience of the precipitation from the cloud in which the precipitation originates to the ground.

with the ceiling height, rainfall intensity, and previous precipitation experience of the air in the low levels above the ground for studying the descent experience, listed as variable six in Table 4.

Figs. 2, 3, and 4 show the data for the three winter storms meeting the large-scale uplift conditions. The individual effects have already been discussed.(1)

In summary, the data show that in each case the ground-level Sr^{90} concentration follows closely the trends in the height of the ceiling. A poorer correlation is noted for the average precipitation rate, and very little correlation is noted for the influence of previous precipitation experience at the generating level.

Many observers (5,6,7) note that the Sr^{90} concentration decreases during the progress of a storm. We observed that the Sr^{90} concentration decreased with a gradual lowering of the ceiling. In the two cases where precipitation continued with a rise of the ceiling, a corresponding increase in the Sr^{90} concentration was observed. The ceiling is an index of the humidity profile and depth of the layer of air in which most of the evaporation occurs. Thus, the descent experience of falling precipitation is considered to be an important parameter in determining the variations in Sr^{90} concentrations in ground-level precipitation from layer clouds formed by large-scale uplift of stable air. The descent experience may be conveniently expressed by ceiling height measurements.

Preliminary data for convective storms

Collections of precipitation from convective storms have been made since March 1961. Although the meteorological and radiochemical analyses are not yet completed, the data indicate that the proximity of the cloud tops to the jet stream, where mixing between the stratosphere and the troposphere occurs, and the extent of the vertical development of the clouds determine the Sr^{90} concentration in rain reaching the ground.(8) For the 7 showers collected during this period, analysis is being made of the peak Sr^{90} concentration (dpm/l) during precipitation as a function of the maximum height of the cloud tops during the storm and the distance of these clouds from the jet axis. Variations are seen in the individual showers. The largest Sr^{90} concentrations are in the rain which falls after the cloud has achieved its greatest vertical development. Showers which occur further from the jet show lower concentrations but still with variations due to the changing height of the precipitation generating level.

The preliminary data are given in Table 5. Fig. 5 shows these data superimposed on a contour plot, taken from Giles(9), of the Sr^{90} concentrations (dpm/10^3 **S.C.F.**) in the atmosphere with respect to the tropopauses and the jet axis. This plot has been normalized to correspond to an idealized jet stream model. Two observations are apparent. The peak activity

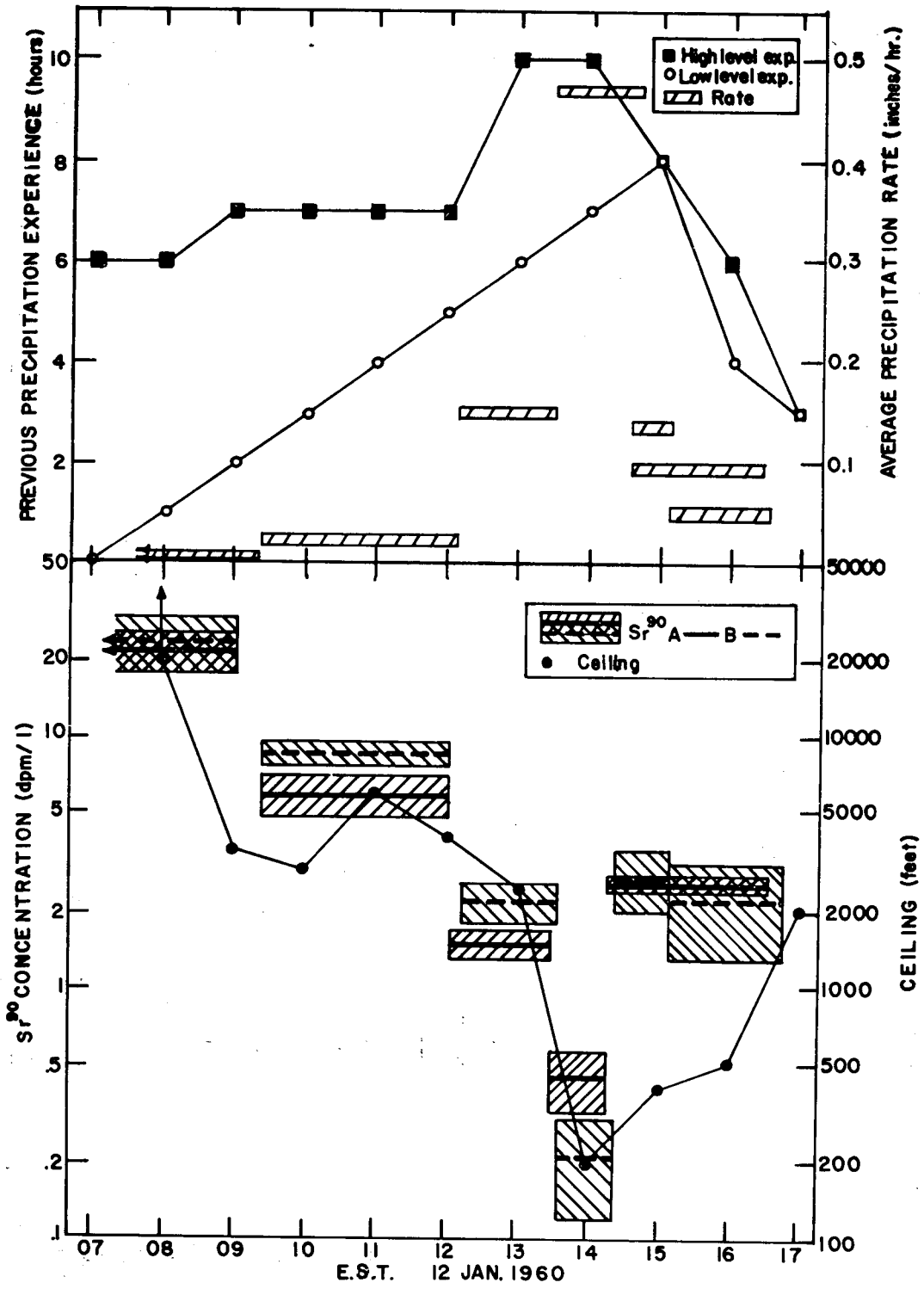

Fig. 2

Sr^{90} Concentration and Meteorological Data for Storm No. 1 - 12 January 1960

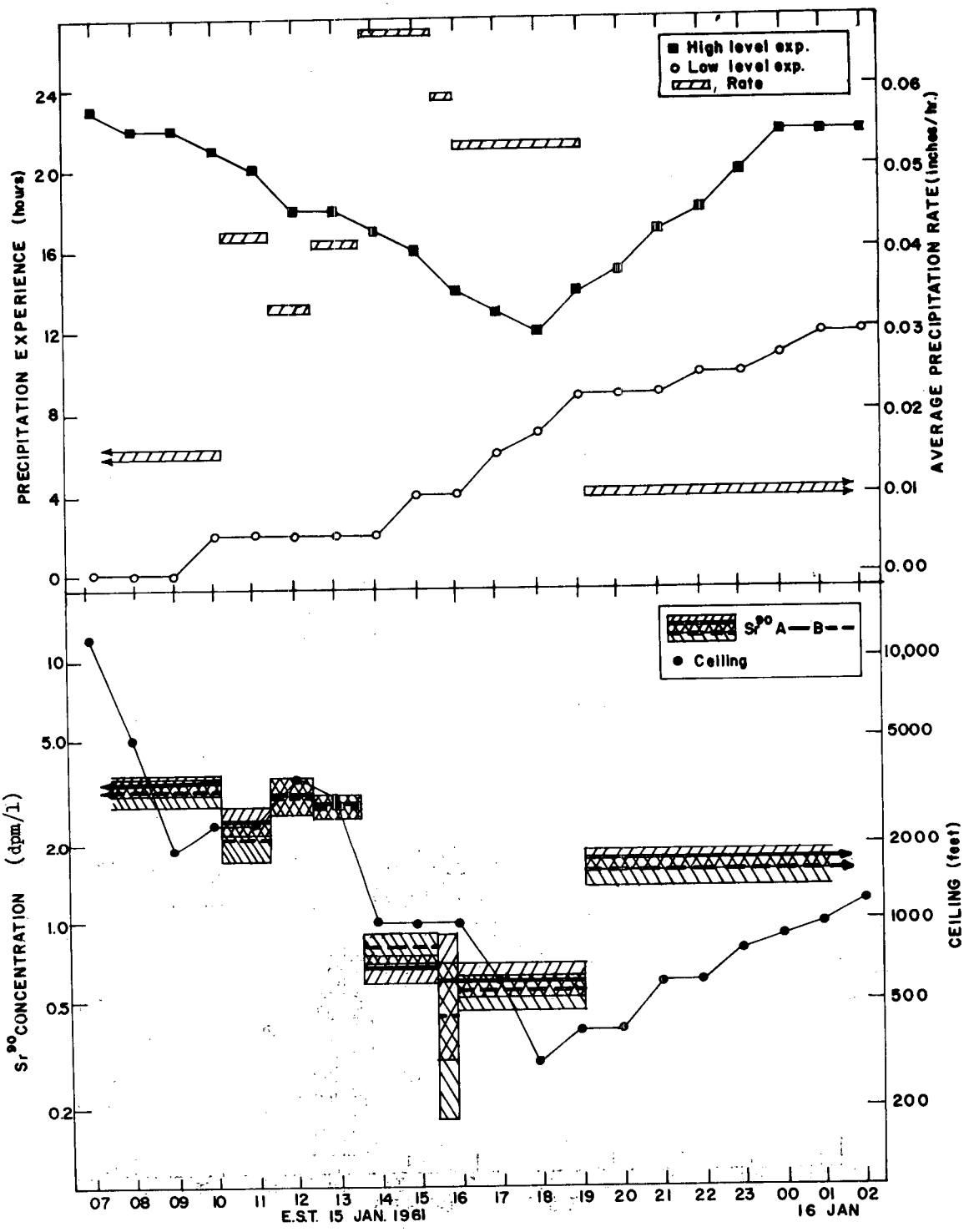

Fig. 3

Sr^{90} Concentration and Meteorological
Data for Storm No. 3 - 13 January 1961

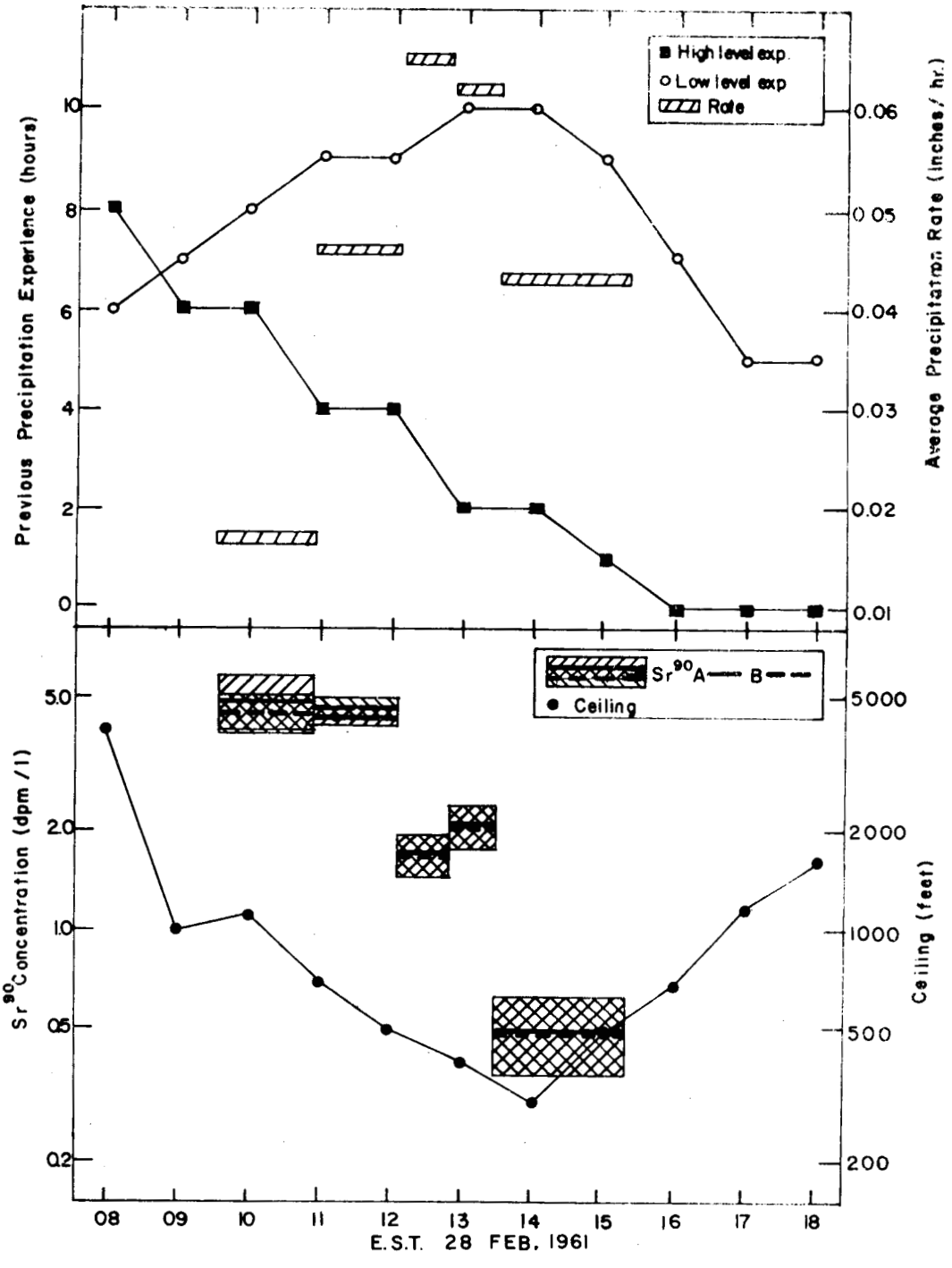

Fig. 4

Sr^{90} Concentration and Meteorological
Data for Storm No. 4 - 28 February 1961

TABLE 5

Preliminary Data for Convective Storm Studies

Date (1961)	Volume (liters)	Total Sr^{90} (dpm)	Average Sr^{90} Conc. (dpm/l)	Peak Sr^{90} Conc. (dpm/l)	Height of Cloud Tops Relative to Tropopause (+) feet above (-) feet below	Distance from Jet Axis (+) miles north (-) miles south
March 8	27.3	240	8.8	51.0	+5,500	+75
May 8	15.1	5.9	0.39	0.4	-21,000	-400
May 8	45.3	278	6.1	6.7	-1,000	-400
May 9	44.2	248	5.6	9.9	-1,000	-150
July 24	78.1	43.1	0.55	1.1	-4,500	-300
August 25	85.5	30.2	0.35	0.8	-4,000	-190
September 14	51.8	10.8	0.21	1.6	-11,500	-450

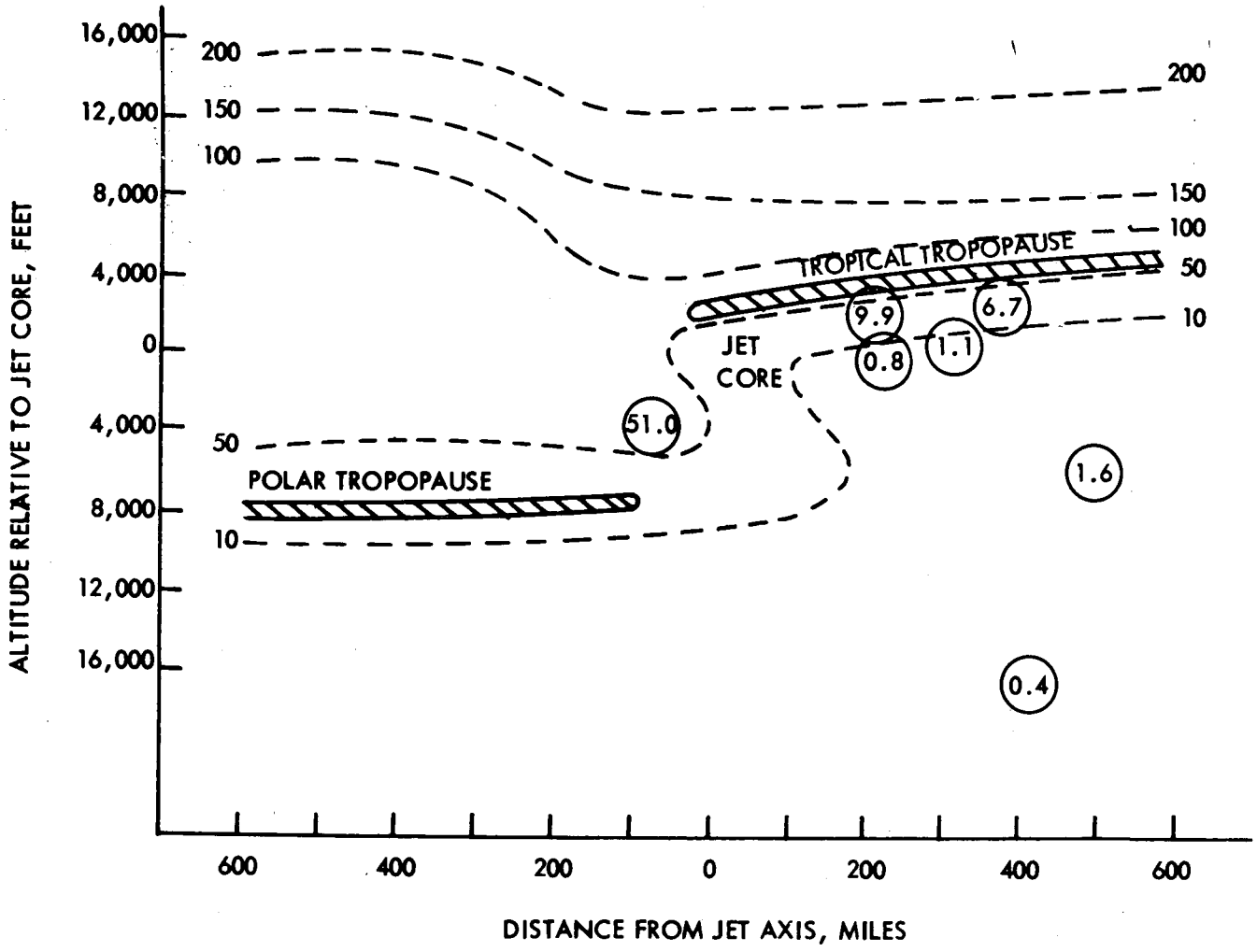

Figure 5 - Peak Sr^{90} Concentration (dpm/l) in Convective Shower Precipitation Plotted at Relative Position of Maximum Cloud Tops and Distance from Jet Core. Data are Superimposed on Air Concentration Plot (dpm/1000 scf) from Giles (HASL-115).

concentration in each convective shower appears to be strongly a function of the two assumed variables; and, although no correlation is inferred between airborne concentation in dpm/10^3 S.C.F. and precipitation in dpm/l, a good over-all fit is observed. A possible dependence on precipitation concentration with respect to the Sr^{90} "reservoir" available for nucleation both below and above the tropopause is suggested. Our highest observed concentration value of 51 dpm/l, which occurred in the March 8, 1961 rain, is accompanied by our observation that the cloud system itself penetrated through the tropopause break in the vicinity of the jet. The possibility that penetration into the stratosphere by convective systems may be a significant alternate method of transport of stratospheric debris into the troposphere is now being examined. Penetration of convective storms into the stratosphere is most probable during the spring months and may be coincident with a larger-scale downward flow through the break in the tropopause in the vicinity of the jet.

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RADIONUCLIDES IN PRECIPITATION

Philip W. Krey and Alan Walton
Isotopes, Incorporated
Westwood, New Jersey

A great deal of attention has been focused on the monitoring of Sr^{90} in world-wide fallout. This interest is wholly justified because of the hazards which would be attendant to the widespread dissemination of large quantities of this radionuclide. However, there are other radionuclides produced in nuclear detonations which have been the subject of a great deal less study but which may ultimately reveal the basic principles governing the transport of material in the upper atmosphere and the deposition of world-wide fallout. Once these principles are clearly understood and a concise mechanism describing world-wide fallout developed, then the hazards associated with large and small nuclear detonations can be better evaluated.

Isotopes, Inc. has maintained a precipitation monitoring station at its laboratories in Westwood, N.J. for the U.S. since February 1958. In addition to the usual Sr^{90} assay of each precipitation sample, Sr^{89} and Ba^{140} determinations have also been performed until the levels fell below our detectable limits in 1959. With the resumption of the U.S.S.R. atmospheric testing, however, our monitoring program for these and other short-lived nuclides has been reactivated. Fig. 1 presents a summary of the barium data in terms of its activity ratio to that of Sr^{90}.

Ba^{140} has a 12.8 day half-life which makes it an excellent indicator of debris from a recent nuclear detonation. For most large yield bursts, the Ba^{140}/Sr^{90} ratio at shot time is approximately 1200 derived primarily from the 14 Mev neutron fission of U^{238}(1). There would be little fractionation expected between Sr^{90} and Ba^{140} because of the similarities in the half-lives of their rare gaseous precursors and also because of the chemical similarities of the 90 and 140 mass chains.

The data in Fig. 1 show the arrival and behavior of debris from at least 3 and possibly 4 weapons test series from 1958 through early 1959. The dashed lines represent the theoretical radioactive decay of the ratio Ba^{140}/Sr^{90} drawn through the experimental points. When extrapolated to the production value of 1200, this ratio should identify the major source of the radioactive debris. Therefore, in April and May 1958, fallout from the U.S.S.R. series in February is evident. From July through September of that same year, debris from the combined United Kingdom and the U. S. HARDTACK series can be identified. From October 1958 through February 1959, much of the radioactive fallout can be assigned to the U.S.S.R. series in October 1958 and possibly a small amount to the United Kingdom tests in September 1958.

In essentially all of these test series, a number of large yield detonations took place. In reality then, a single production date of radioactive debris from any one series is unrealistic. However, a hypothetical mean shot date can be estimated which is intended to represent the weighted average date of all the high yield shots in any one series. These mean shot dates are indicated in Fig. 1 by small arrows at each of the test series. In all cases the extrapolated line reaches the 1200 ratio earlier in time than the estimated mean shot date. This is the result of residual Sr^{90} remaining in the atmosphere from previous nuclear tests which reduces the Ba^{140}/Sr^{90} ratio and gives rise to the apparent earlier shot date.

It is possible to estimate the contribution of each of these test series to the total Sr^{90} fallout during the months immediately following the tests from the mean shot dates and the observed Ba^{140}/Sr^{90} ratios. During the spring of 1958, the U.S.S.R. tests in February contributed about 25% of the total Sr^{90} fallout in precipitation samples at Westwood, N.J. In the summer and early fall of 1958, the combination of the United Kingdom April tests and the HARDTACK series contributed about 75% of the total Sr^{90} being deposited. (Since both of these test series took place near the same times and in the same general latitudes, a mean shot date of June 7 was used for the combined United Kingdom and U. S. HARDTACK series.) If the reasonable assumption is made that all the Ba^{140} in precipitation at Westwood during the early months of 1959 was derived from the U.S.S.R. series in October 1958, then approximately 60% of the total Sr^{90} fallout at this time was due to these U.S.S.R. tests. By and large, a similar pattern in Ba^{140}/Sr^{90} ratios was observed by Martell at Bedford, Mass.(2).

A similar and corroboratory pattern is shown in Fig. 2 for the Sr^{89}/Sr^{90} ratios in precipitation samples collected during the same period. The dashed lines represent the theoretical radioactive decay of the ratio normalized to the experimental points. The identification of fallout with 3 nuclear test series is again evident. The scatter of the points is much greater in early 1958 than in later periods probably because the U.S.S.R. tests in February did not allow sufficient time to permit adequate mixing of the U.S.S.R. debris with earlier debris.

The effect of the United Kingdom series in September 1958 was evident in the Ba^{140}/Sr^{90} ratios, but appears to be more clearly presented in the Sr^{89}/Sr^{90} ratios. However, additional discussion will be brought to bear on this subject when the Ba^{140}/Sr^{89} ratios are considered. The U.S.S.R. debris becomes dominant in mid-February and prevails until September 1959. By assuming a zero time Sr^{89}/Sr^{90} ratio of 160 and the same mean shot date for each test series as was assumed earlier, one can again calculate the percentage of the total Sr^{90} fallout contributed by each series during the months immediately following the series. These calculations agree with the estimates derived from the Ba^{140}/Sr^{90} ratio, namely 25% from the U.S.S.R. February test in the spring of 1958 and 75% from the combined United Kingdom spring and U.S. HARDTACK tests in the summer of that year.

The Sr^{89}/Sr^{90} ratios also permit a little more fine structure to be resolved during the latter months of 1958 than did the Ba^{140}/Sr^{90} ratios. From the third week of October to the end of December, the Sr^{89}/Sr^{90} ratios indicate that about 30% of the Sr^{90} fallout was contributed by the U.S.S.R. October 1958 tests. From January through mid-February 1959, the Sr^{90} contribution from these tests increased to about 60%. From the middle of February through August of 1959, the U.S.S.R. October 1958 series contributed about 75% of the Sr^{90} being deposited.

The Ba^{140}/Sr^{89} ratios are presented in Fig. 3 where the dashed line once again represents the theoretical radioactive decay of the ratio. Working backwards in time, one can discern that the major Ba^{140} and Sr^{89} activities in late October 1958 and subsequent months were derived principally from the U.S.S.R. tests in October and little or none from the United Kingdom tests in September. Therefore, the indications of the Ba^{140}/Sr^{90} and Sr^{89}/Sr^{90} ratios to the contrary, the United Kingdom tests in September contributed little Sr^{90} to the fallout at Westwood during late fall and early winter of 1958 and 1959. The Sr^{89} and Ba^{140} from the U.S.S.R. October shots were diluted with Sr^{90} from previous tests (probably HARDTACK) making the Sr^{89}/Sr^{90} and Ba^{150}/Sr^{90} ratios appear to be of United Kingdom origin.

Another item of interest is that the 9 darkened points in Fig. 3 that appear to be too high all represent dry samples or rains of very small magnitude ($\leq$0.02 in.). A theory which could explain this phenomenon is that the Sr^{89} with a 2.3-min. Kr^{89} rare gaseous precursor would tend to concentrate on small particles more so than the Ba^{140} with a 16 sec. Xe^{140} rare gaseous precursor. In dry or periods of very light rain, it would be reasonable to expect the larger particles to deposit in the precipitation collectors more readily than the smaller particles which could give rise to the observed effect. In deference to some of the previous papers at this symposium which assigned the bulk of the residual radioactivity in the stratosphere to submicron particles, it is important to recall that the periods under consideration in this report are a few months after nuclear weapons testing when larger particles still remain suspended.

The very early arrival of the October debris strongly suggests that it was of tropospheric origin. On the other hand, the smooth behavior of the ratio for 3½ months is equally a strong indication that the debris falling out through the entire period was of stratospheric origin. A compromise may be struck such that the tropospheric debris of recent vintage was from the same shot or shots producing the major fraction of the stratospheric fallout of Ba^{140} and Sr^{89} in subsequent months, thereby producing no change in the Ba^{140}/Sr^{90} ratio. Meteorological data about the winds in the upper troposphere would have been very helpful in testing these thoughts. It is unfortunate that the Ba^{140} activities became undetectable in mid-February of 1959 because the last 5 points intimate that still another change was taking place.

Referring again to Fig. 2, it can be seen that at mid-February the Sr^{89}/Sr^{90} ratio first reached the decay that it subsequently followed for the next 5 months. This was the beginning of the spring "peak" in fallout which has been observed annually for the last 8 years. It is recognized by many that this "peak" is due to a net downward mixing of stratospheric air and its injection into the troposphere during late winter and early spring. (2,3,4) One might conclude then that the fallout from the third week in October 1958 through January 1959 was derived primarily from the upper reaches of the troposphere and the lower extremeties of the stratosphere. From February to October 1959 the downward mixing of the stratosphere yielded a homogenized source of fallout from greater heights.

The extrapolated Ba^{140}/Sr^{89} ratio in the summer of 1958 intercepts the production value of 7.5 on June 20. There were a number of HARDTACK shots in this vicinity so that the Ba^{140} and Sr^{89} activities in the debris might be derived primarily from these few shots. If this were the case, then the contribution of the combined United Kingdom and U.S. HARDTACK series to the Sr^{90} fallout during the summer as estimated earlier is in error. This should be substituted by a sole HARDTACK contribution of about 50%. Moreover, if the fallout was from the few shots, then the rapid arrival intimates tropospheric debris. Here again the value of meteorological data during sample collection times is apparent. The Ba^{140}/Sr^{89} ratios for the spring of 1958 and for the fall and winter of 1958-1959 give excellent correlation with the mean shot dates of the two U.S.S.R. series intercepting the production value at February 26 and October 17, respectively.

The behavior of these nuclide ratios offer an opportunity for some additional speculation. During periods in which the ratios exhibit a fairly smooth decay, it follows that the atmospheric source giving rise to this fallout must have a relatively uniform composition. First of all, with the exception of the U.S.S.R. October 1958 tests, there was an interval of on the order of one month or more between the beginning of a nuclear test series and the arrival of the peak in the ratios at Westwood. This time delay suggests that the debris was of stratospheric origin. Secondly, the duration and consistency of the periods of decay are greater

than one might expect from a tropospheric source. Thirdly, as was described earlier, the spring "peak" in fallout is due to a net downward mixing of stratospheric air and its injection into the troposphere during late winter and early spring. Therefore, the source of relatively uniform composition giving rise to the fallout in the springs of 1958 and 1959 was probably the lower stratosphere. The origin of the fallout during the end of 1958 and the beginning of 1959 has already been suggested as the lower stratosphere in which the relative composition changed slightly with the advent of spring. This is not to say that the stratosphere is homogeneous; but that there is a relatively rapid movement of debris initially after a test series to yield a metastable distribution within the stratosphere. Project HASP (High Altitude Sampling Program of the Defense Atomic Support Agency) has shown that the distribution of debris in the stratosphere can have very steep gradients (4). The rainfall data suggests that after the initial formation of these distributions, subsequent variations in them occur at a much slower rate producing an apparent fallout source of relatively uniform composition.

Such concepts are admittedly broad generalizations of the true situation. The experimental points from which these thoughts were developed show enough scatter to illustrate that there are short-term variations in the parameters governing radioactive fallout in rain. The W^{185} analyses in precipitation shown in Fig. 4 offer additional evidence of this nature.

Several tungsten isotopes were generated in a number of shots from the U.S. HARDTACK series. The tracers are unique for this series since they were never generated to any significant level by any other nuclear tests. The plot in Fig. 4 was constructed by adding the activities in each rain within overlapping 30-day intervals and dividing by the summation of rain in the interval. The interval was then advanced in 10-day increments and the process repeated. All the W^{185} data were corrected for radioactive decay to the arbitrary date of August 15, 1958. The Sr^{90} data computed in the same way are presented for comparison. We believe that the first two pulses in the concentrations of W^{185} in precipitation samples from mid-1958 to the end of October 1958 reflect the shot schedule of the devices generating the tungsten. From mid-November through June 1959, the W^{185} concentrations mirror the Sr^{90} concentrations, once again attesting to the stratospheric origin of the fallout. In Fig. 5, the Ba^{140}/W^{185} ratio is plotted as a function of time. The ratio fluctuates somewhat in May and June 1958 but then reaches a maximum in early July. Subsequently the ratio falls off at a steeper rate than the theoretical decay (dashed line) indicating the incursion of stratospheric air richer in W^{185}. The behavior of W^{185} in rain samples collected in the middle latitudes until mid-September 1958 does not represent the behavior of the total HARDTACK W^{185} that was generated in the HARDTACK series. Consequently, any radioactive material balance calculations made in the summer of 1958 using the total W^{185} HARDTACK production value are necessarily in error.

In early October, the Ba^{140} levels from the U.S.S.R. shots begin to build up even greater than the W^{185} levels from HARDTACK shots such that the over-all ratio increases. By late October a curious situation develops in which the ratio apparently decays at the theoretical rate but concurrently along two parallel lines. This would suggest that there are two different sources of fallout with relatively uniform composition that intermittently release debris for incorporation into the rains at Westwood. Martell (3) has observed a similar phenomenon in the Ba^{140}/Sr^{90} ratio at Bedford, Mass. in July through September 1958. This he explained on the basis of polar tropospheric air poorer in Ba^{140} concentrations being the source of rains yielding the lower Ba^{140}/Sr^{90} ratios. Whatever the actual case may be, the Ba^{140}/W^{185} ratios demonstrate that while the source of the fallout during the periods under consideration in this report exhibits a general character of stability, there are discreet deviations.

It has been often repeated that the fate of the radioactive debris injected into the stratosphere is greatly influenced by when and where the injection takes place. A point in case is the high altitude rocket shot, Orange at Operation HARDTACK. Rh^{102} was produced in this shot and serves to identify Orange debris as uniquely as did the W^{185} for previous HARDTACK shots. The first appearance of Rh^{102} in monthly precipitation samples at Westwood was in October 1960, two years after the detonation took place. In Fig. 6 the behavior of the Rh^{102} concentration in rain samples can be seen, demonstrating that the debris from the Orange shot is finally descending from its lofty residence in the stratosphere and mixing with tropospheric air at increasing rates. A continuing study of the distribution of this radionuclide, both in the stratosphere and precipitation samples, offers great promise for investigation of particle transport problems at very high altitudes.

During the current U.S.S.R. testing, an air sampling program has been instituted at Westwood using the same air filtering system employed by Lockhart at NRL. Twenty four hour samples are collected on Chemical Corps type 6 paper representing a filtered volume of about 42,000 S.C.F. of air. These samples are allowed to decay for two hours after which they are counted on a 3 in. x 3 in. NaI(Tl) crystal of a multi-channel gamma ray spectrometer. By this time, the Ra^{222} daughters have decayed away to small levels and the Rn^{220} (thoron) daughter products can be subtracted by normalizing their standard spectra against the Tl^{208} 2.6 Mev gamma ray. This approach eliminates contributions of the natural radioactive products in air and minute influxes of fission debris can be rapidly identified. Fig. 7 gives a record of these air concentrations since the first identification of recent debris on September 16, 1961.

Many of these samples are further assayed by gamma ray spectrometry during the weeks following collection to permit resolution of the spectra into Ba^{140}, Zr^{95}, Ru^{102}, and I^{131} activities. The ratios of some of these nuclides will permit the identification and possibly the

approximate contribution of the shots producing the fallout. In addition, a small disk is punched out of some of the samples for beta decay measurements which will also serve to identify the shots giving rise to this airborne activity. Specific attention will be given to the air filter samples before, during and after a rain in an effort to correlate the concentrations of some of the nuclides, particularly I^{131}, in air and in the rain. Unfortunately, the data are not available for presentation at this time. However, a few general observations can be made.

1. Based on preliminary Mo^{99}/Ba^{140} ratios, shot identification of the first few peaks in air concentrations have been made. The preliminary shot dates which are indicated in Fig. 7 next to the samples from which identification was derived are probably accurate to within 2 days. The data show the arrival of the first debris in about 6 days after the shot. Although all the shots are conducted at this time were air bursts to the best of our knowledge, the rapid transport time of the debris strongly indicates the debris to be of tropospheric origin. Furthermore, the weather pattern in the upper troposphere depicted by the U.S. Weather Bureau supports this rapid arrival of debris along the east coast of the United States from the shots around September 10.

2. The total beta decay measurements also show the arrival of fresh debris from recent shots as indicated in Fig. 7.

3. Preliminary data suggest that the I^{131} concentrations in the air filters are slightly low indicating that there is either some fractionation between the Mo^{99} and I^{131} or that the filter paper is preferentially permitting some of the I^{131} to pass through, as I_2 perhaps.

4. For the full interpretation of the experimental data being collected, the local meteorology at the collection station should be known. Consideration should be given to what meteorological measurements are practical and meaningful and a common measurement plan adopted by all the individual rain monitoring stations of the AEC.

In summary, nuclide ratios in precipitation can be used to identify the source of radioactive fallout and to approximate the contribution of certain test series to the total Sr^{90} fallout at specific times. The rainfall data suggest that the debris from nuclear detonations initially has a relatively rapid movement within the stratosphere to yield a metastable distribution which subsequently changes at a much slower rate. More meteorological data should be gathered at the fallout collection sites to assist in the interpretation of the fallout measurements.

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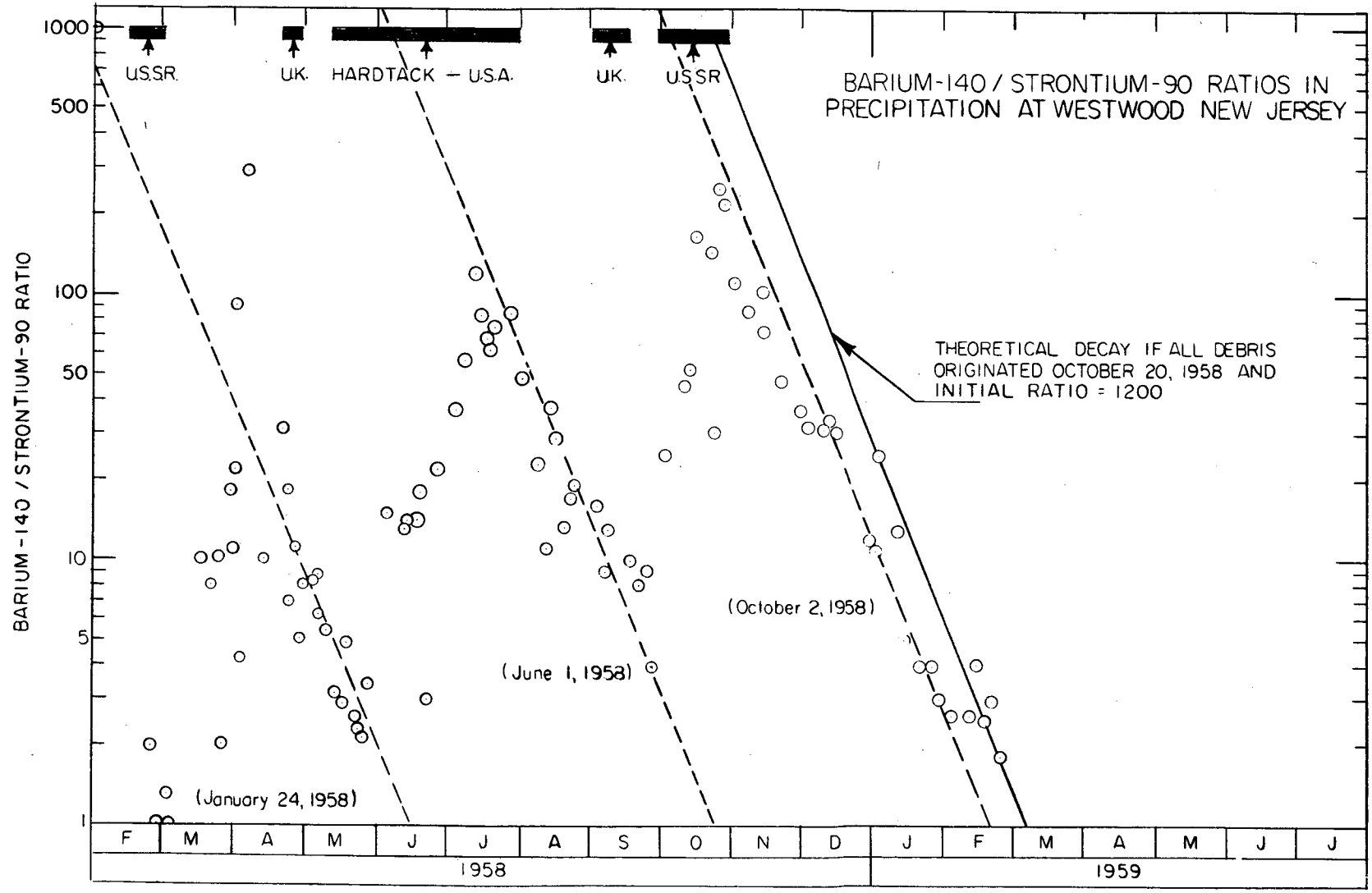

BARIUM-140 / STRONTIUM-90 RATIOS IN PRECIPITATION AT WESTWOOD NEW JERSEY
BARIUM-140 / STRONTIUM-90 RATIO
1000
500
100
50
10
5
1
U.S.S.R.
U.K.
HARDTACK — U.S.A.
U.K.
U.S.S.R
THEORETICAL DECAY IF ALL DEBRIS ORIGINATED OCTOBER 20, 1958 AND INITIAL RATIO = 1200
(January 24, 1958)
(June 1, 1958)
(October 2, 1958)
F M A M J J A S O N D J F M A M J J
1958
1959

Figure 1

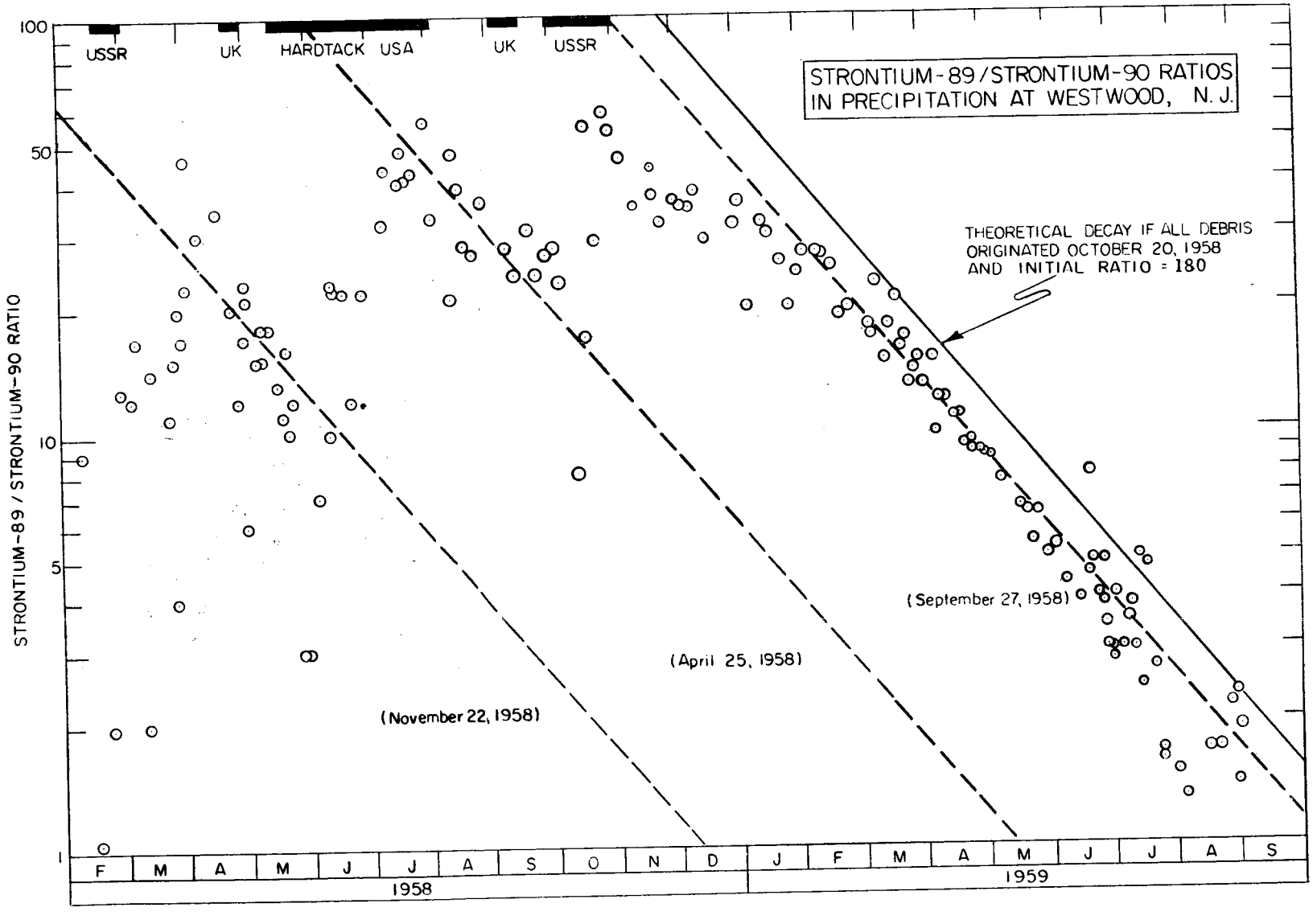

STRONTIUM-89/STRONTIUM-90 RATIOS
IN PRECIPITATION AT WESTWOOD, N.J.
STRONTIUM-89 / STRONTIUM-90 RATIO
100
50
10
5
1
USSR
UK
HARDTACK USA
UK
USSR
THEORETICAL DECAY IF ALL DEBRIS
ORIGINATED OCTOBER 20, 1958
AND INITIAL RATIO = 180
(September 27, 1958)
(April 25, 1958)
(November 22, 1958)
F M A M J J A S O N D J F M A M J J A S
1958
1959

Figure 2

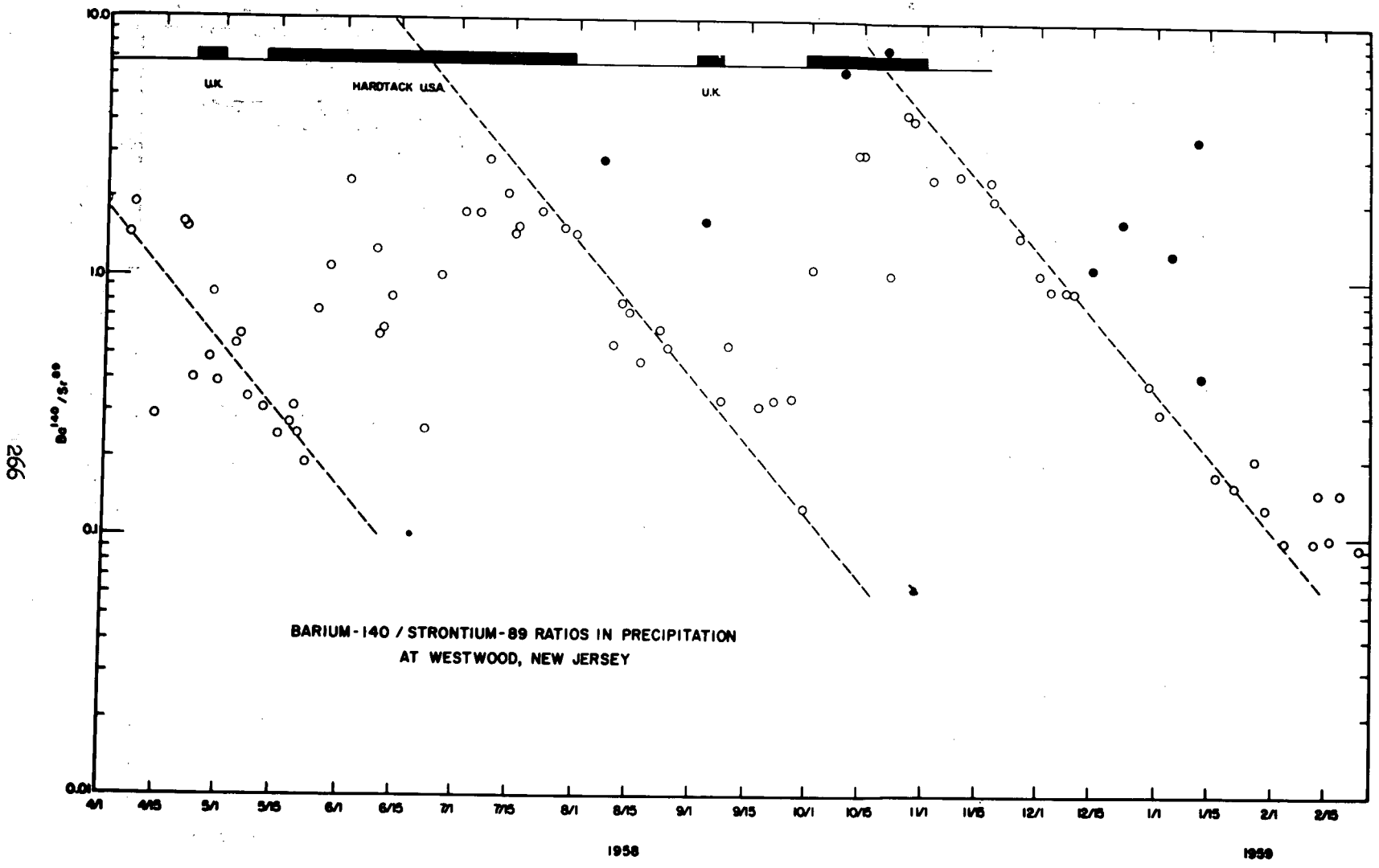

10.0
1.0
0.1
0.01
Ba^{140}/Sr^{89}
U.K.
HARDTACK U.S.A.
U.K.
BARIUM-140 / STRONTIUM-89 RATIOS IN PRECIPITATION
AT WESTWOOD, NEW JERSEY
4/1
4/15
5/1
5/15
6/1
6/15
7/1
7/15
8/1
8/15
9/1
9/15
10/1
10/15
11/1
11/15
12/1
12/15
1/1
1/15
2/1
2/15
1958
1959

Figure 3

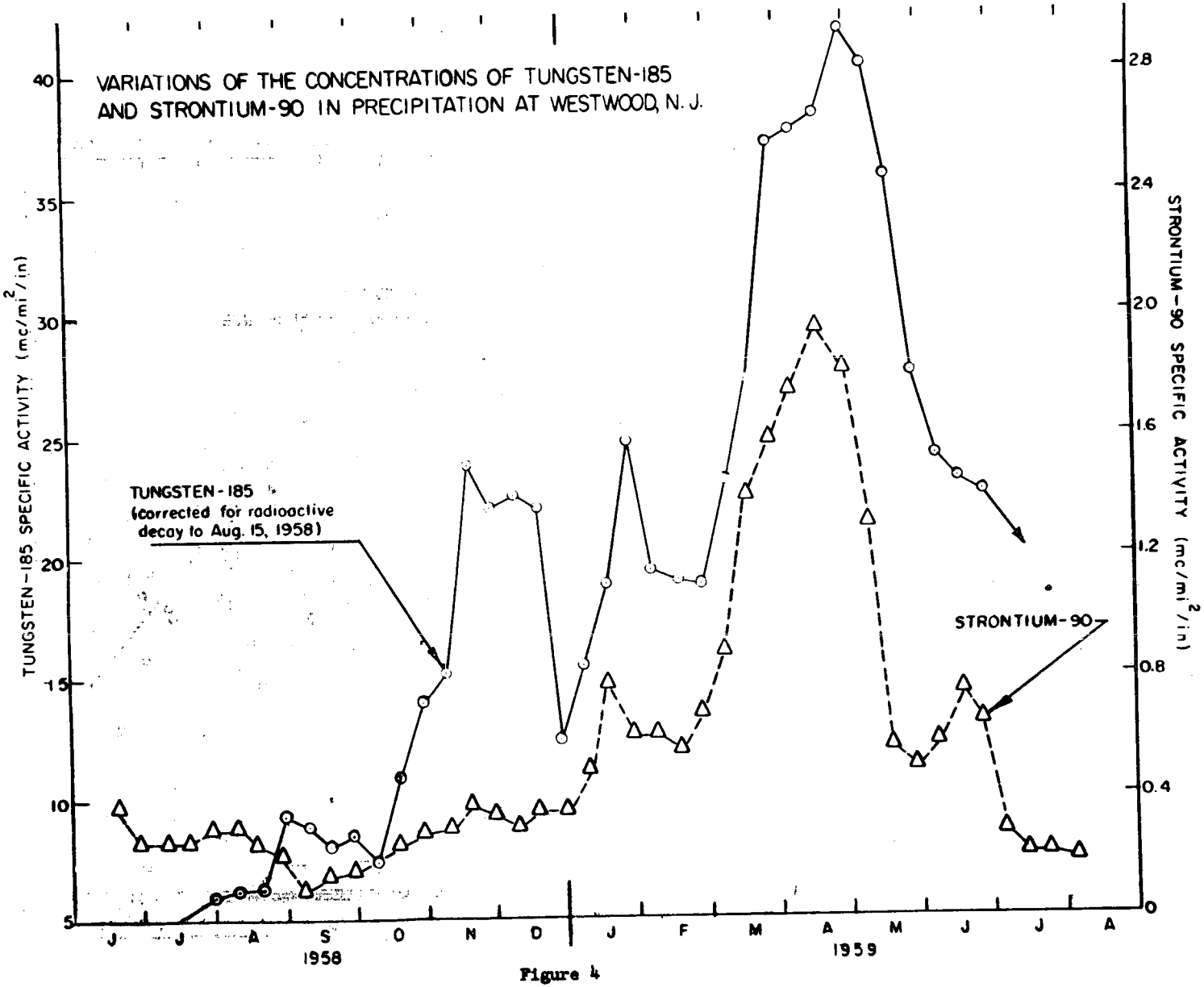

VARIATIONS OF THE CONCENTRATIONS OF TUNGSTEN-185
AND STRONTIUM-90 IN PRECIPITATION AT WESTWOOD, N. J.
TUNGSTEN-185 SPECIFIC ACTIVITY (mc/mi²/in)
STRONTIUM-90 SPECIFIC ACTIVITY (mc/mi²/in)
TUNGSTEN-185
(corrected for radioactive
decay to Aug. 15, 1958)
STRONTIUM-90
40
35
30
25
20
15
10
5
2.8
2.4
2.0
1.6
1.2
0.8
0.4
0
J J A S O N D J F M A M J J A
1958
1959

Figure 4

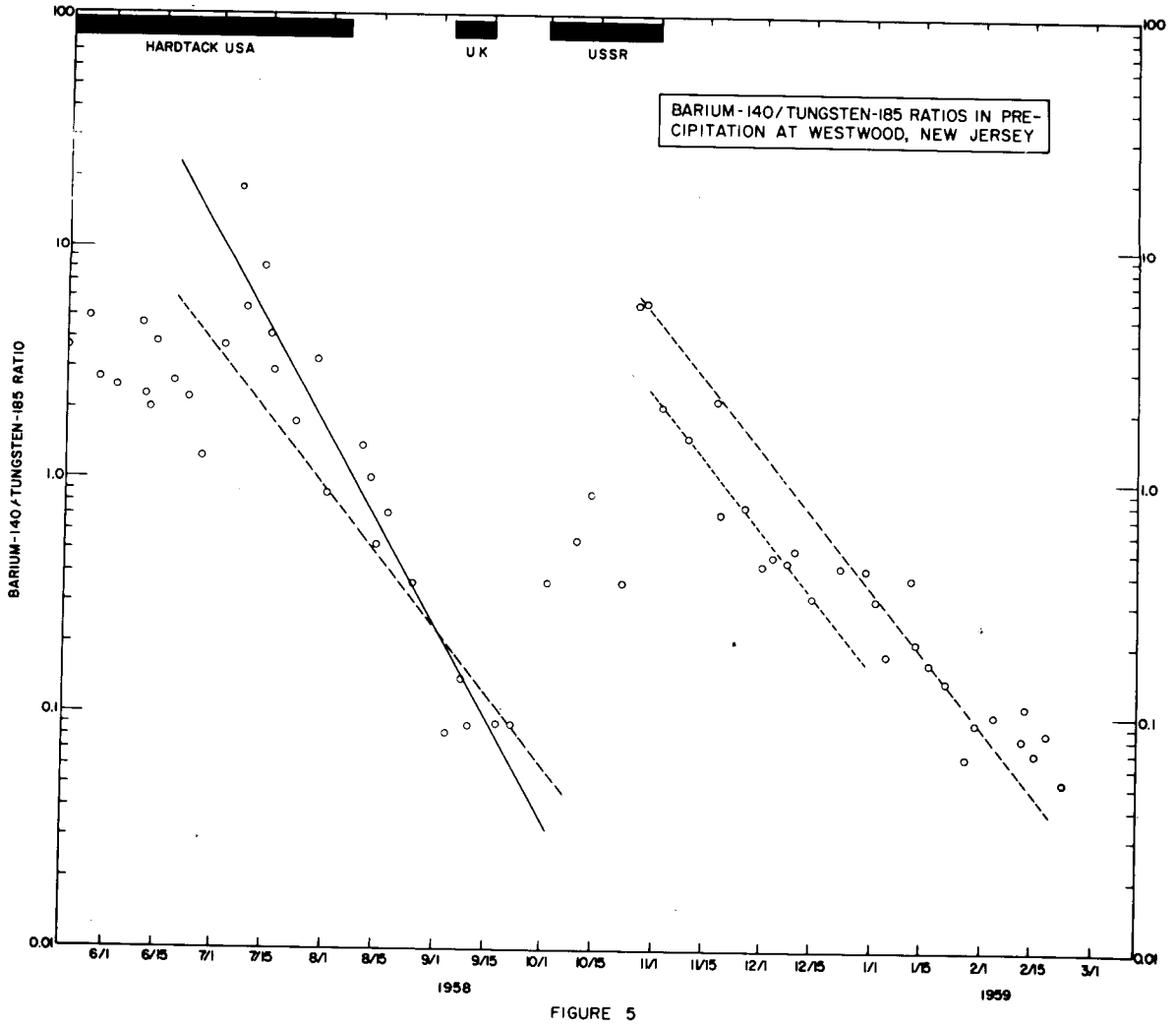

HARDTACK USA
UK
USSR
BARIUM-140/TUNGSTEN-185 RATIOS IN PRECIPITATION AT WESTWOOD, NEW JERSEY
BARIUM-140/TUNGSTEN-185 RATIO
100
10
1.0
0.1
0.01
6/1
6/15
7/1
7/15
8/1
8/15
9/1
9/15
10/1
10/15
11/1
11/15
12/1
12/15
1/1
1/15
2/1
2/15
3/1
1958
1959

FIGURE 5

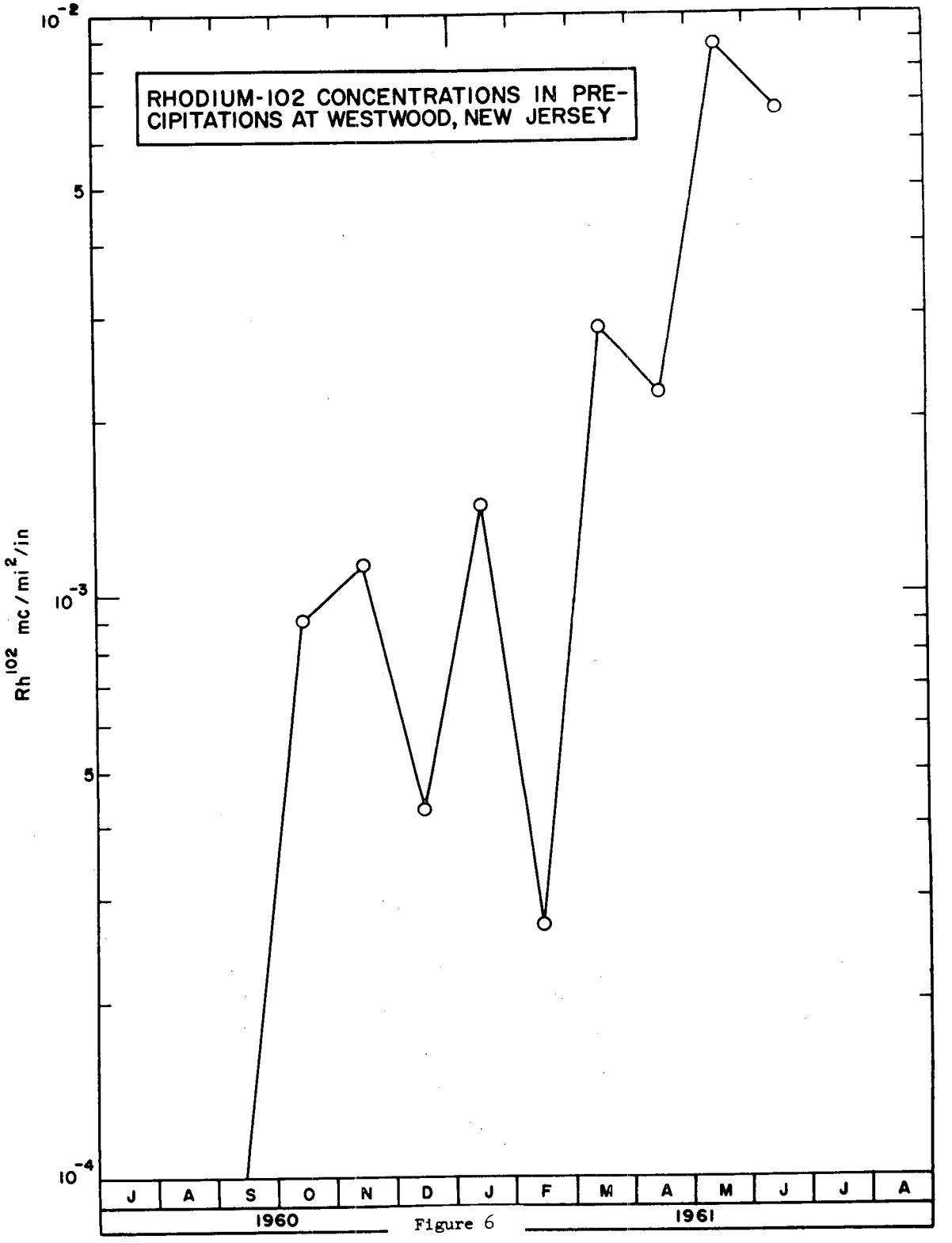

RHODIUM-102 CONCENTRATIONS IN PRECIPITATIONS AT WESTWOOD, NEW JERSEY
10^{-2}
5
10^{-3}
5
10^{-4}
Rh^{102} mc / mi^2 /in
J A S O N D J F M A M J J A
1960
1961

Figure 6

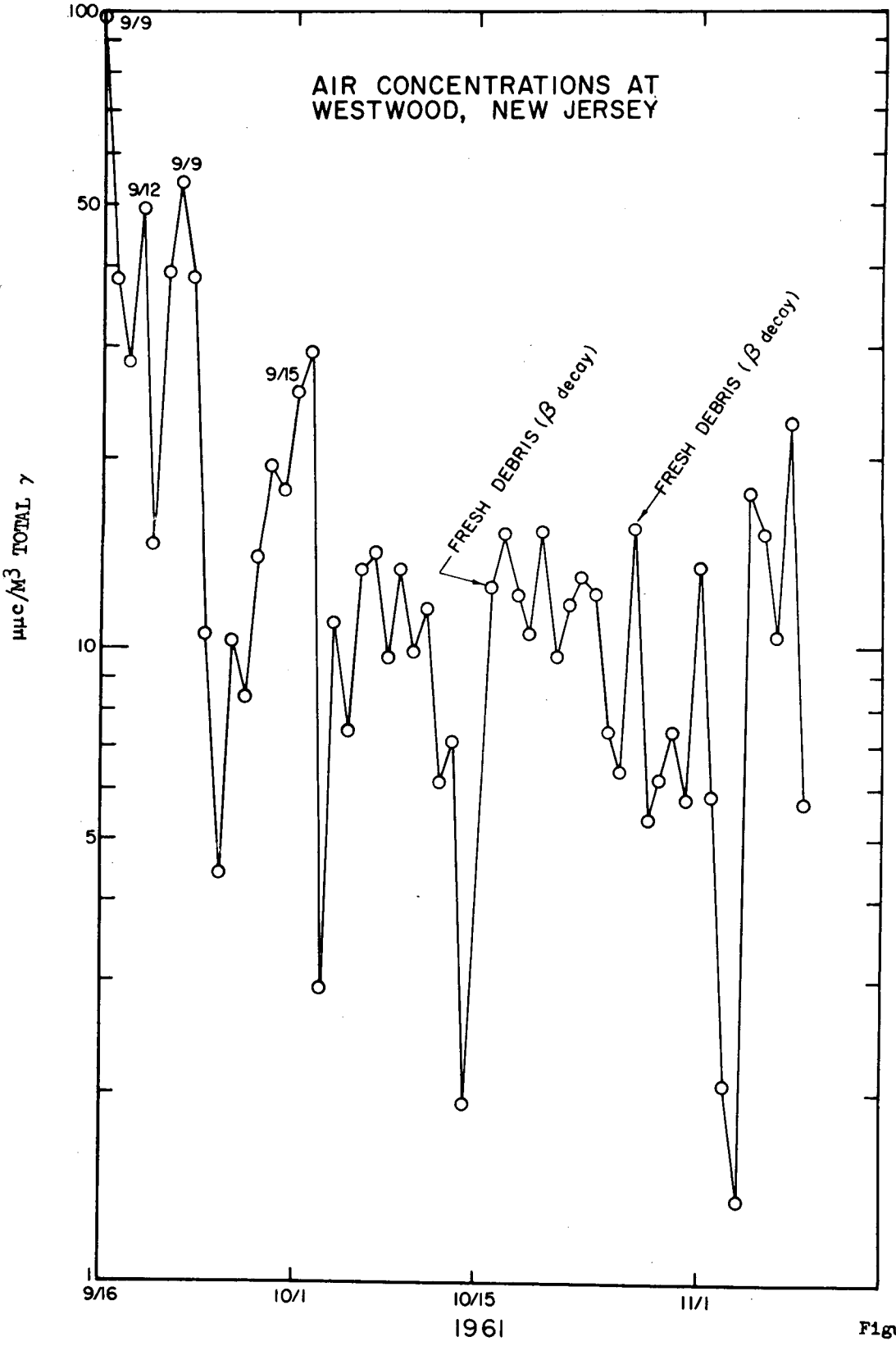

AIR CONCENTRATIONS AT
WESTWOOD, NEW JERSEY
μμc/M3 TOTAL γ
100
50
10
5
1
9/9
9/12
9/9
9/15
FRESH DEBRIS (β decay)
FRESH DEBRIS (β decay)
9/16
10/1
10/15
11/1
1961

Figure 7

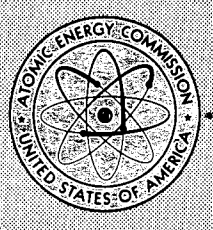
TID-7632

RADIOACTIVE FALLOUT FROM NUCLEAR WEAPONS TESTS

Proceedings of a Conference Held in Germantown, Maryland

November 15-17, 1961

BOOK 2



United States Atomic Energy Commission
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TID-7632 (Book 2)
BIOLOGY AND MEDICINE

RADIOACTIVE FALLOUT FROM NUCLEAR WEAPONS TESTS

PROCEEDINGS OF A CONFERENCE

NOVEMBER 15-17, 1961

Held in the

U. S. Atomic Energy Commission Auditorium

Germantown, Maryland

Under the auspices of
Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission
Washington 25, D. C.

Edited by

Alfred W. Klement, Jr.

February 1962

MEASURED AND PREDICTED CONTRIBUTIONS FROM FALLOUT TO ENVIRONMENTAL RADIATION LEVELS

William R. Collins, Jr.
Health and Safety Laboratory
U. S. Atomic Energy Commission
New York, New York

Concentrations of Sr^{89} and Sr^{90} have been measured in fallout collections in New York City since the middle of 1954. In addition, during the period of intense nuclear weapons testing that occurred in 1958 and the moratorium of testing that followed in 1959, determinations of Cs^{137}, W^{185}, Zr^{95}, Ru^{106}, Ce^{144}, and Y^{91} were performed whenever possible at the same site. These data have been previously reported (1,2,3, 4) and have proven useful first, as a means of estimating the contributions made by fallout to environmental radiation levels and second, as a means of tracing the atmospheric history of fallout in New York from different origins. Currently they offer a basis for predicting what effects the 1961 U.S.S.R. test series will have on future fallout levels.

Calculated infinte plane gamma dose rates resulting from cumulative levels of Zr^{95}, Ru^{106}, Cs^{137}, and Ce^{144} that were deposited in New York during 1958 and 1959 have been reported.(4) Considering only the 2-year fallout increment, it was found that the dose rates from fallout reached as high as 50% of total natural terrestrial and cosmic radiation dose rates. Even with maximum corrections for weathering, scattering, and shielding, total open field dose rates from fallout appeared to be at least 0.2 μrad/hr. through most of the observation period with from 65 to 95% of the total affect attributable to Zr^{95} and Cs^{137} activities.

Although direct measurements of the gamma emitters in fallout in New York City prior to 1958 are not available, the similarities in the half-lives of Sr^{89} and Zr^{95}, and Sr^{90} and Cs^{137} suggest the use of the earlier radiostrontium measurements(1,2) for the estimation of depositions

of the two major contributors to fallout gamma radiation doses prior to 1958. The theoretical Cs^{137} Sr^{90} and Zr^{95} Sr^{89} radioactivity production ratios for thermonuclear weapons have been estimated to be 3.1 and 1.54, respectively.(5) Over the two year period the former ratio would not be expected to change appreciably, while the latter could range from 3.1 to 1.5 in mixtures of debris up to 14 months old. The 1958 and 1959 data yielded average monthly Cs^{137} Sr^{90} and Zr^{95} Sr^{89} ratios of 1.5 ± 0.49 and 2.5 ± 1.3, respectively. Thus, with the assumption that fallout conditions from 1955 through 1957 were not radically different from those during 1958 and 1959, multiplication of the early Sr^{89} concentration by 2.5 and the Sr^{90} concentration by 1.5 should give reasonable estimates of the Zr^{95} and Cs^{137} depositions.

Infinite plane gamma radiation doses calculated from cumulative Zr^{95} and Cs^{137} levels obtained from these conversions are illustrated in Fig. 1. No corrections are made for weathering, shielding, or scattering. Zr^{95} and Nb^{95} are assumed to be in transient equilibrium at a ratio of 1.0 to 2.4. Values plotted after June 1959, when Sr^{89} depositions were below the limits of detection, are obtained by correcting the cumulative Zr^{95} level at that time for monthly decay.

The New York dose rate curves are in fair agreement with gamma radioactivity assays of soils and ionization chamber measurements of dose rates at several sites of similar latitudes.(5) Assuming an average per cent contribution to the Cs^{137} and Zr^{95} total of 20% for other gamma radioactivities and an overall physical reduction factor of 1/10, it is seen that total open field gamma radiation dose rates from fallout reached as high as 0.3 and 0.6 μrad/hr. in 1955 and 1959, respectively and remained at values in excess of 0.1 μrad/hr for most of the 1955 - 1960 period.

If it is assumed that the peaks in the Zr^{95} dose rate curve are attributable to the dispersion of debris from individual test series to New York, then more or less discrete contributions from Operation TEAPOT in 1955, and the U.S.S.R. test series and Operation REDWING in the first half of 1956 are discernible. However, quantitative resolutions of the mixed depositions that occurred during this period and through the middle of 1958 would be difficult using either isotopic or nuclide ratio data. The unique production of radio-tungsten in the U. S. Operation HARDTACK I series and the preponderance of debris from the U.S.S.R 1958 autumn tests over earlier debris during the first half of 1959 permit reasonable calculation of the compositions of monthly fallout depositions in New York after May 1958 through analysis of Sr^{89} to Sr^{90} ratios.

This analysis is illustrated in Fig. 2. The ratios plotted are corrected for HARDTACK I contributions using theoretical Sr^{90}/W^{185} and Sr^{89}/W^{185} radioactivity production ratios of 0.00380 and 0.436, respectively, as of June 1, 1958. The relative contributions from the U.S.S.R. series in late 1957, early 1958 and late 1958 are then calculated from

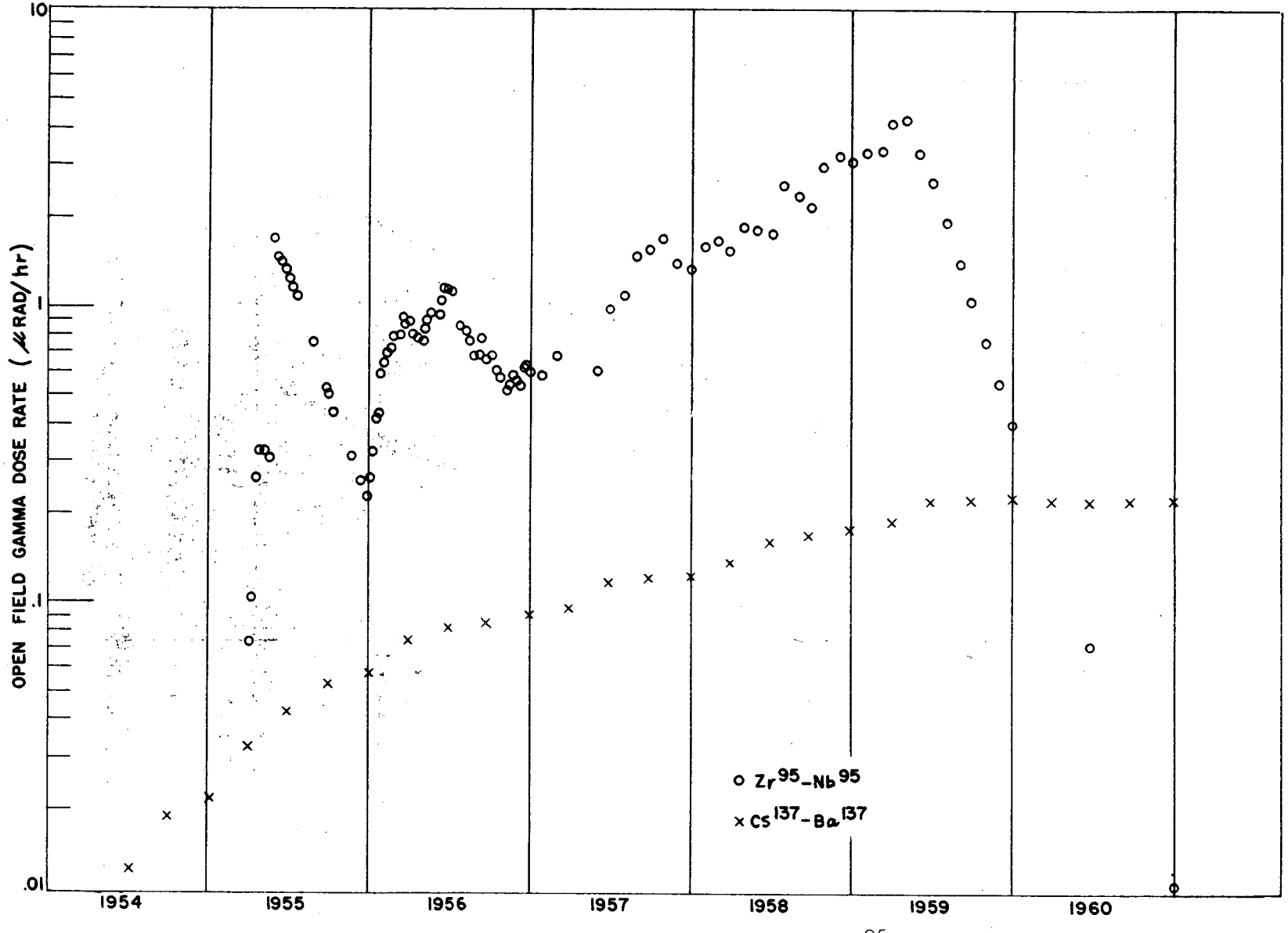

FIGURE 1 Estimated Contributions from Cumulative Levels of Zr^{95} and Cs^{137} to Infinite Plane Gamma Dose Rates in New York City

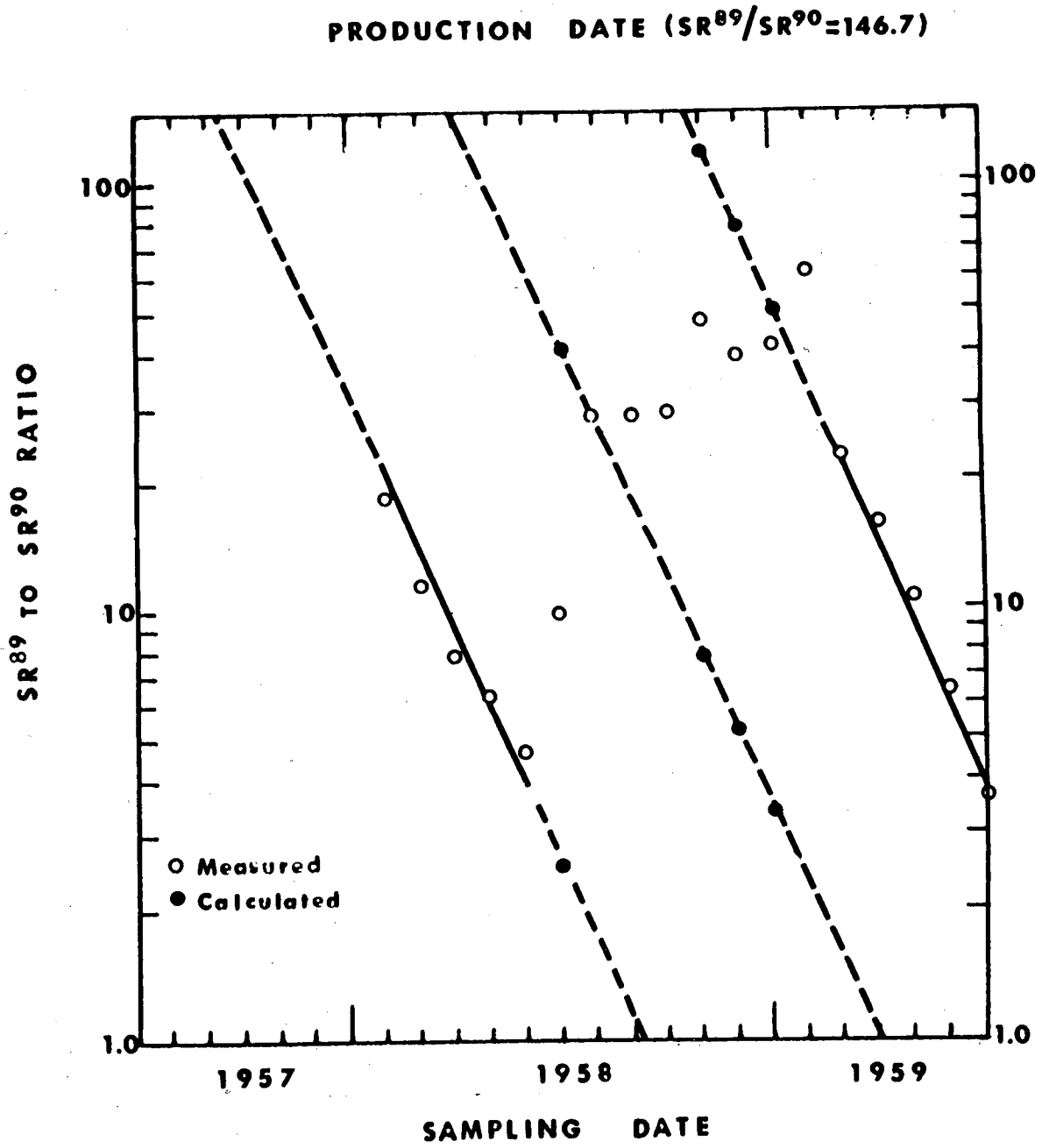

Figure 2. Sr^{89}/Sr^{90} Ratios from Non-HARDTACK I Debris in New York City during 1958 and 1959.

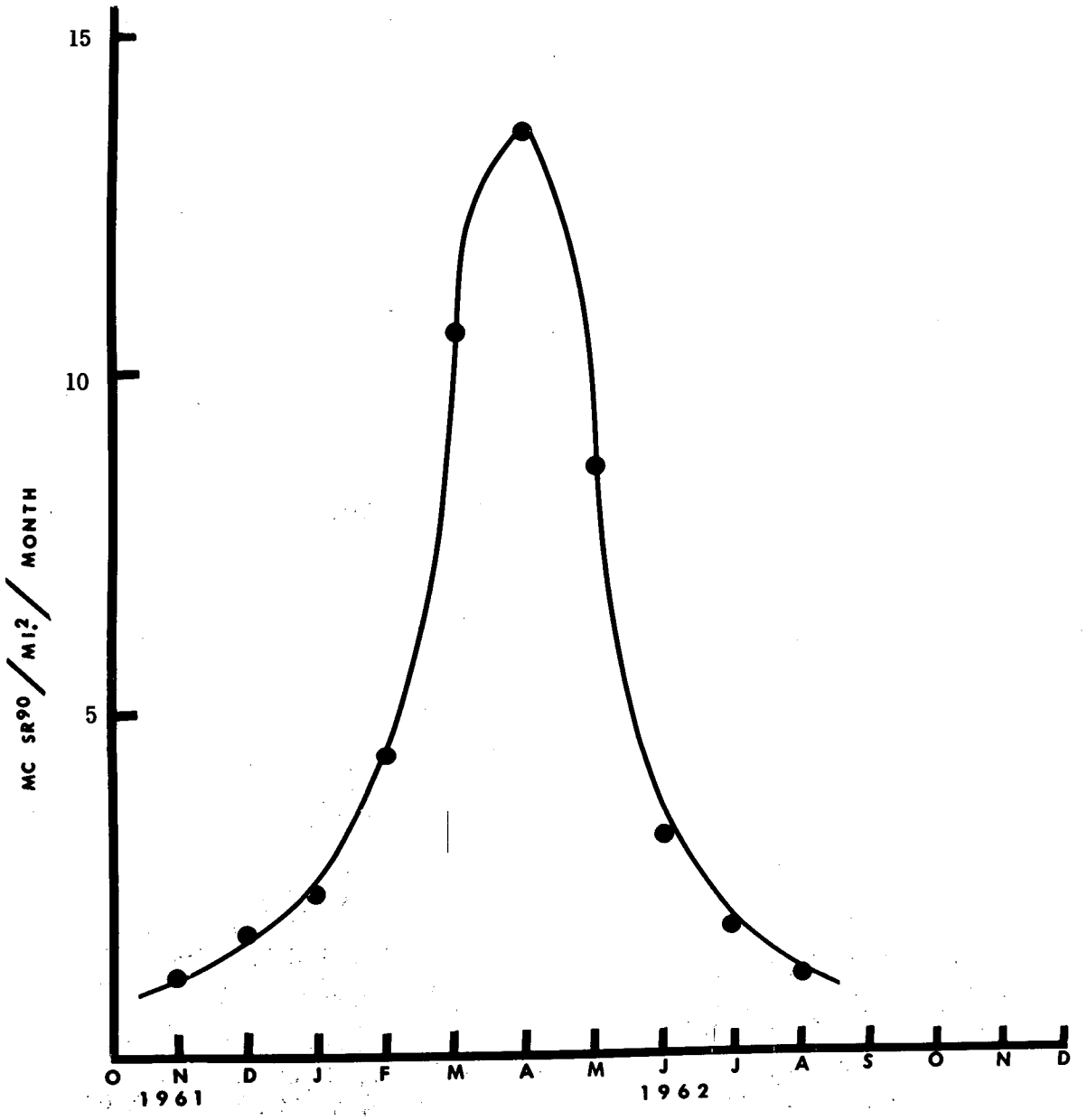

Figure 3. Expected Fallout Rate of Sr^{90} in New York City from the 1961 Soviet Test Series.

the position of the points in respect to lines drawn from the midpoint of each series with the theoretical slope of the Sr^{89}/Sr^{90} ratio decrease with time.

The results of these calculations are listed in Table 1. Although it is not possible to trace HARDTACK I radioactivities beyond July 1959, the abundance of U.S.S.R. debris in New York fallout prior to that time is clear. Moreover, the rapid dissipation of the arctic debris as compared to the gradual decline in equatorial radioactivities is established.

The similarity of the timing and locations of the U.S.S.R. 1958 and 1961 test series leads to the extension of these observations to late 1961 and early 1962 fallout predictions. With the assumptions that differences in the spacing of the tests within the series and in the altitudes of the detonations were not great, calculated specific fission fractions for the 1958 series (mc $Sr^{90}/mi.^2/mo./in.$ rain/MT of fission equivalent) should obtain to the 1961 series. Then with the additional assumptions that 1962 rainfall will conform to the 40-yr. average monthly rainfalls and that the fission yield of the 1961 test series was 25 MT, estimates of monthly depositions in New York during 1962 are obtained.

These values are illustrated in Fig. 3. As in 1958, the peak depositions should occur in the spring months and may reach as high as 13 mc $Sr^{90}/mi.^2/mo.$ by April. If altitude, test schedules, or other factors were significantly different the peak depositions may be delayed but between October 1961 and October 1962 the total addition to the present Sr^{90} inventory should be about 47 $mc/mi.^2$ If Zr^{95} and Cs^{137} dispersions are consistent with those for Sr^{90} the infinite plane dose rate from the Zr^{95}-Nb^{95} system alone will be about 9 μrads/hr. at the time of the peak Sr^{90} deposition and a 30-yr. dose of about 38 and 45 mrads will be produced by the year's depositions of Zr^{95} and Cs^{137}, respectively.

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1. Health and Safety Laboratory. Environmental contamination from weapons tests. U. S. Atomic Energy Commission, New York, report HASL-42, October 1948. 359pp.

2. Hardy, P., Jr., and S. Klein. Strontium program summary reports. HASL-55, 1959; HASL-65, 1959; HASL-69, 1959; HASL-77, 1960; HASL-84, 1960.

3. Welford, G. A., and W. R. Collins, Jr. Fallout in New York City during 1958. Science 131:1711-1715(1960).

TABLE 1

Sources of Sr^{90} fallout in New York City during 1958 and 1959

Sampling Month	Sr^{90} Level (mc/mi.^{2})		
	Tests Between Oct. 1958 and Apr. 1959	Operation HARDTACK I	U.S.S.R. Oct. 1958
May 1958	3.44	0.006	
June 1958	1.23	0.039	
July 1958	1.25	0.224	
Aug. 1958	0.358	0.195	
Sept. 1958	0.378	0.197	
Oct. 1958	0.596	0.227	0.347
Nov. 1958	0.450	0.320	0.420
Dec. 1958	0.119	0.179	0.474
Jan. 1959		0.521	0.479
Feb. 1959		0.512	1.30
March 1959		0.707	3.74
April 1959		0.492	3.67
May 1959		0.313	1.21
June 1959		0.260	2.17
July 1959		0.104	0.416

4. Collins, W.R., Jr., G. A. Welford, and R. S. Morse. Fallout from 1957 and 1958 nuclear test series. Science 134:980(1961).

5. Hallden, N. A., I. M. Fisenne, L.D.Y. Ong, and J. H. Harley. Radioactive decay of weapons debris. U. S.AEC, Health and Safety Laboratory Technical Memorandum, 61-20, New York, 1961, 5pp.

6. Gustafson, P.F. Assessment of radiation dose due to fallout. Radiology 75:282-287(1960). Vennart, J. Increase in local gamma-rag background due to nuclear bomb. Nature 185:722-724(1960). Pierson, D. H., and L. Salman. Gamma-radiation from deposited fallout. Nature 184:1678-1679(1959).

DISCUSSION

Mr. Joshua Z. Holland, Chairman of the session, directed the discussion of the papers presented in the session which follows.

NEWELL: Dr. KRUGER, I should like to know the variation in rainfall rates at the time you were measuring the ceiling. Is there an indication of an inverse relationship?

KRUGER: During the presentation of my paper last night, I showed the Sr^{90} concentration as a function of three variables, of which the intensity of rainfall was one. I didn't get a chance to show that the reason we think the ceiling is so important is that in each case the changes in Sr^{90} concentration correlated well with changes in the ceiling. We did not observe a good correlation with the hours of experience at the generating level of rainfall, or with the intensity of the rainfall itself. For the latter, in two cases we did get a nice correlation. However, in the third case we got one exactly opposite: the more intense the rainfall, the greater the Sr^{90} concentration.

MACHTA: We intend to collect some rainfall on the top of the RCA building in New York during a period of very dry weather and see whether the concentration of Sr^{90} in water will actually increase because of the evapororation that takes place with the falling droplets.

DINGLE: I wanted to direct a question to Dr. NEWELL. This migration of ozone to the mid-latitude low point was quite interesting. I was wondering if Dr. NEWELL would give us some comments on his views of how this gets into the troposphere.

NEWELL: That is something I haven't dealt with in this particular paper, but after my reading on this matter, I would be in agreement with BREWER, STALEY, and others, who have argued that its exchange is along surfaces in which potential temperature is conserved. All the evidence that Dr. LOCKHART supplied yesterday argues in favor of this, because his peaks in the surface concentration move north and south with the season, and these peaks are to the south of the tropopause gap. The fission products diverge in the lower temperature from this region. So we are in agreement on that point. But as to whether there is an increase in the tropospheric - stratospheric exchange in the spring, this is another question which I would not wish to answer at this time.

HOLLAND: Dr. MARTELL and others have expressed ideas on this. I wonder if we couldn't have comments on this question. Dr. NEWELL has mentioned a way in which the fission products can, and probably do, come out of the stratosphere. My question is: Is this by far the predominant mechanism? Is it a dominant one? Is it one of many? I would like to hear from Dr. MACHTA or Dr. MARTELL.

MACHTA: In our office we haven't come to any conclusions as to the exact mechanism. We certainly believe that one one of the methods of exchange is through the tropopause gap. There was a weak suggestion that the October 1958 Russian debris never got far enough south as to exit through the gap and may have left through the tropopause in the polar regions. I hope we will have enough observations during the forthcoming months to demonstrate that this did or did not occur, and I think we have a golden opportunity to see whether or not polar air must reach the gap in order to exit into the troposphere.

MARTELL: I think that both the Paetzold ozone data and the Ba^{140}/Sr^{90} ratio data that my associates and I obtained in 1958, give a very clear indication that mixing of stratospheric air into the troposphere takes place not just at mid-latitudes in the region of the tropopause break but at higher latitudes as well. We distinguish with the Ba^{140}/Sr^{90} ratio data, for example, a difference in nuclide ratio of stratospheric material in the polar troposphere and subtropical troposphere, which is maintained for period of several months. There is apparently no mixing of aerosols between the two regions, or at least aerosol scavenging in frontal rains seems to minimize mixing effects. This implies that downward mixing at latitudes north of the main tropopause break is a general phenomenon or that mixing takes place through secondary breaks at higher latitudes.

HOLLAND: Prof. STALEY is here. I wonder is he would care to comment on this question. Prof. STALEY of the University of Arizona.

Dean O. STALEY: I feel that this transfer or exchange can occur readily at all latitudes north of something like 30° N, or latitudes in which we have extratropical disturbances, and that the "breaks" can appear in association with specific disturbances. An exchange is typically associated with each disturbance in which the air which enters the troposphere does not immediately return to the stratosphere, so that the debris can remain in the troposphere long enough to experience scavenging. Now, most of this exchange will take place in a preferred latitude, say 35° N, or in the region where on mean charts the tropopause gap is most readily imposed. But I think you have to keep in mind that the tropopause is not a rigid barrier with a hole in it at the gap region which allows mass exchange only at that latitude. The break may come or go at all latitudes, and this does occur primarily at, say, 35° N or S; and so there is a temptation to impose a "gap" on a mean meridional cross section. I might add that this past spring we sampled with a small aircraft for total beta activity below, within, and above dry baroclinic zones in the troposphere. Preliminary counting shows maximum activity within the zone, which supports the concept of entry into the troposphere within sloping dry baroclinic layers which occur in association with extratropical

disturbances. I think there is a lot more uncertainty, at least in my mind, concerning the spring maximum of exchange. I think that the moving up and down of the tropopause throughout the year must be an important factor here, but there are other viewpoints on this. In the spring months the tropopause is moving upward. The maount of mass in the stratosphere is decreasing in high latitudes at this time of year, so that the region of exchange is moving upward toward the level of high concentrations andthe exchange involves higher concentrations in the lower stratosphere than it does in the fall months, when the tropopause is moving downward. I am sure that there are other factors which are important here, such as the subsidence associated with the late winter explosive stratospheric warming as piinted out by Libby and Palmer. I would not say that I am convinced that upward movement of the tropopause is the only explanation. I think this must be an important factor.

DINGLE: I would like to ask Dr. STALEY if in his studies of the tropopause behavior he had indications of actual deterioration of the tropopause as a barrier during this period of migration, so that it would not be such a barrier to stratosphere-troposphere exchange.

STALEY: You ask about whether there would be deterioration in the tropopause itself during the periods of upward and downward migration?

DINGLE: Yes, whether during the spring, in upward migration, you actually had a sort of deterioration and then regeneration of the tropopause periodically as it progresses upward; this kind of thing.

STALEY: No, I don't think so. I don't think there is any particular deterioration of the tropopause at that time. There appears to be a rather large number of sizeable exchanges that occur at this time of the year; as evidenced, I think, through a large number of intense baroclinic zones in the high troposphere, very dry zones, which is consistent with the idea of their having recently subsided from high levels. There, of course, can be found in the fall and in the winter, and not so readily in the summer. But, of course, in the summer months the number of intense extratropical disturbances goes down, too. I think my answer would be that there is no general deterioration of the intensity of the tropopause during the spring or the fall. However, in the spring there seems to be a greater number of these exchanges, for reasons that I don't understand.

KULP: I would like to comment on the source of introduction of the debris. I think that the distribution in terms of Sr^{90} in rain and the distribution of Sr^{90} on the ground is such that the dominant mechanism has to be introduction in the latitude zone of 30-50°N. If the introduction was primarily in the polar regions directly into the troposphere, then we should see a sharp gradient of deposited Sr^{90} decreasing from 70°N to 30°N. We do not find that. Rather, we find that 30 to 40°N and 40 to 50°N it is very similar, and even 50 to 60°N in the same rainfall belts it is similar. I think a second evidence is that the concentration of Sr^{90} in rain or in ground level air decreased northward about 60°N.

I also think I can recall some tropospheric aircraft sampling, including some by Sweden, which showed that going north actually showed a modest decrease rather than an increase in the concentration of Sr^{90} in the air. The picture is slightly confused by the introduction of debris directly into the polar troposphere by Russian testing but when this is taken into account it is clear that the dominant portion of the stratospheric debris is transferred in the temperate rather than the polar latitudes.

LANGHAM: I am concerned about the reports by Kirk Lidén of Sweden that the Laplanders in Sweden and Norway are now running cesium body burdens that are from 30 to 50 times higher than we have ever found, even in the vicinity of 35 to 40°N. I am curious to know if there is any meteorological explanation as to why, in the Arctic Circle, we have this very perculiar case where people are showing up with cesium burdens that are astronomically high compared to what is found elsewhere.

HOLLAND: Before I ask the panel to comment on this, I think it is only fair to ask you whether you find it impossible to account for this on biospheric or ecological gounds.

LANGHAM: We can make some explanation on that basis, but I am not satisfied that it is enough. In orher words, it seems that it can not all be accounted for by the fact that they live on reindeer meatand goat cheese, even though these are both browsing type animals. If, indeed, the radioactivity always falls off toward the poles as beautifully as the curves indicate, it is hard to think that concentrations in the northerly latitudes are high enough to make even goat cheese impalatable.

HOLLAND: Do any of the speakers wish to speakto this?

MARTELL: I would like to make one comment with respect to the specific activity of rains versus latitude. My associate, Dr. P. J. DREVINSKY, has been looking at the concentrations of individual nuclides in rains and snow, including some from 77°N. The concentrations of these Greenland snows averaged 5 to 10 times the maximum concentrations observed in North Temperate latitudes. I think that the marked difference in fallout deposition versus latitude is due in large part to differences in precipitation versus latitude, so that the air concentration of the polar troposphere is apparently similar to that of the mid-latitude troposphere, if not higher.

KRUGER: I would like to point out that we have had some discussions in our own group which stem from some indications from thundersotrms which have had tremendous vertical development and may have penetrated into the stratosphere. In our March collection of this year I pointed out that we had a very short period of rainfall in which the Sr^{90} concentration reached a value which we hadn't seen before. However, if this had been looked at by the usual methods of measuring the average Sr^{90} concentration over the whole rain, this high value would have gone undetected. We further believe that the cloud tops from which this precipitation fell may have penetrated into the stratosphere. We intend to continue this research

to see whether such penetration may be an important mechanism for the observed spring peaks.

HOLLAND: Doesn't the seasonal pattern of the fallout differ enough, at least from what we think the seasonal pattern of extremely tall thunderstorm systems is, so that we would question whether this is dominant?

KRUGER: I don't know what percentage this can account for, but we have seen this particular storm and obviously the position of the clouds relative to the jet stream should also be related. Nevertheless, some of these spring thunderstorms do penetrate quite high, and these may be the ones which bring down a larger share of the Sr^{90} than the ones which do not achieve large vertical development.

DANIELSEN: I would like to make several comments. My work on stratospheric-tropospheric exchange processes indicates the outflow process from stratosphere to troposphere is strongly correlated with intense cyclogenesis. As a consequence, stratospheric outflow would be statistically related to the latitude range in which intense vortices are formed, which is going to put in, between 35° and 60° N latitude. Although the radioactivity flows out of the stratosphere in this latitude range it may reach the ground at a wider range of latitudes. It is quite improbable that its descent or trajectory would be straight to the ground. The more probable flow is approximately isentropic, that is, a gradual descent as the air moves southward with the lowest elevation reached some 20° to 25° south of its exit from the stratosphere. At the lower latitude and lower elevation, mixing processes generated by heating at the ground may then bring the radioactivity directly down to the ground as so called "dry fallout". The maximum deposition of dry fallout is expected, therefore, to the south of the point of exit from the stratosphere.

I would also like to comment on another aspect of the stratospheric outflow that is, I think, important to the deposition of radioactivity by rainfall. As the formerly stratospheric air flows southward in the tropopause part of the mass travels a cyclonically curved path. It reaches its lowest elevation at approximately the trough line of the pressure field turns northward, and ascends on the east side of the trough. Now, the air which comes from the stratosphere has, in meteorological terms, particularly high stability, that is, it acts to inhibit small scale vertical mixing. On the east side of the trough the formerly stratospheric air may appear in the troposphere as a thin layer of high stability capping an ascending mass of moist air. The moist air may extend up to the base of the layer, but as long as the stability is present it will inhibit the development of convective cells. With continued ascent of both the dry layer and the moist air it cools less rapidly than the dry air. As a result, the stability in the dry layer is decreased. At some point or points it will be destroyed. Convective cells will rapidly develop, penetrate into the dry air and mix it with the moist air. If the dry layer contains a lot of radioactivity then there is a good possibility that the radioactivity may enter into the precipitation cycle and be brought to the ground.

I think Dr. KRUGER is quite right in saying that the convective cells may penetrate the stratosphere. They may also penetrate the thin layers of formerly stratospheric air. I think, therefore, one has to be very careful in correlating radioactivity with height (or with the tropopause). Because a major portion of the radioactivity may enter the precipitation cycle from a layer at 20,000 ft. in a region where the tropopause is at 40,000 ft.

Finally, if Dr. MANSON is right, that a substantial amount of radioactivity is attached to the sulphate particles in the stratosphere it is conceivable that, during the mixing process just described, the sulphate particles act as nuclei. Drops formed from these nuclei would start out with a high radioactivity content. During their descent the concentration of radioactivity in the drops will increase if there is a loss of mass by evaporation while the heavier radioactive molecules are retained. Roughly speaking, these conditions are more probable if the nucleation occurs at high elevations. The concentration of radioactivity in rainfall may therefore correlate with height parameters although height per se is not physically significant.

HOSLER: I would like to point out that it wasn't height we were correlating, in our paper, with radioactivity. It was the proximity of the top of the radar echo to the area where we thought mixing was occurring in the stratosphere. And it is my feeling at this point that one good reason for the spring maximum is the combination of high instability, maximum instability, leading to very severe storms, with a low tropopause, in the spring which makes it very easy for this debris to come down to the ground. The amount of evaporation which occurs will of course determine how enriched this precipitation is.

HOLLAND: If I might try to merge these last two comments, I think it is of interest that Dr. KRUGER's chart didn't simply show a tropopause in relation to this, but showed a model of the concentration distribution based on air sampling data, which we can call, say, a mean distribution, in relation to these clouds. And Dr. DANIELSEN's comment would be to the effect that it isn't necessarily the mean distribution that matters. It is going to be the distribution at the time of the convection and precipitation, which could depart substantially from this mean distribution. So, a vertical development of precipitation which on the mean chart would not appear to reach the region of high radioactivity, might at that time actually do so.

KREY: We are looking at Be^7/P^{32} ratios in rain with the hope that we might get some verification of this concept that Dr. KRUGER was talking about. If the cloud would get into the stratosphere, we might find a higher Be^7/P^{32} ratio there than with a cloud that didn't get into the stratosphere. This is the result of a more rapid tropospheric washout rate of these cosmic ray produced nuclides permitting a greater equilibrium build up of Be^7 as compared to P^{32} in the stratosphere. We haven't observed

any high ratios that would definitely indicate that such a situation took place, However, this doesn't necessarily prove or disprove anything because of limited data.

MARTELL: First, I would like to correct what is perhaps a misconception. The short-lived Ba^{140} activity which we were discussing was stated to be derived from the stratosphere. We distinguish a difference in Ba^{140}/Sr^{90} ratio of that portion which was transported downward through the polar troposphere as compared to the Ba^{140}/Sr^{90} ratio of that which was transported down at mid-latitudes, presumably through the main tropopause break. We did have other nuclide data, but of course the ratios of other nuclides are not nearly as sensitive as the Ba^{140}/Sr^{90} activity ratios. I don't believe that the difference for Sr^{89}/Sr^{90} would allow you to draw similar conclusions in view of the smaller ratio variations and the analytical uncertainties involved.

DISTRIBUTION AND CYCLING OF FALLOUT NUCLIDES

PART 1. DEPOSITION AND MOVEMENT IN SOILS

Session Chairman: Alfred W. Klement, Jr.
Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission

THE RELATIONSHIP BETWEEN RAINFALL AND STRONTIUM-90 DEPOSITION IN CLALLAM COUNTY, WASHINGTON

Edward P. Hardy, Jr.
U. S. Atomic Energy Commission
Health & Safety Laboratory
New York, New York
and
Lyle T. Alexander
U. S. Department of Agriculture
Soil Survey Laboratory
Beltsville, Maryland

It has been known for some time that rainfall is the primary process by which fission products in the atmosphere are deposited on the surface of the earth. Studies in the United States[1,2] and the United Kingdom[3,4] using open vessel collectors and results of soil analyses[5] have shown excellent correlation between fallout Sr^{90} and the amount of rainfall. Martell[5] demonstrated an increasing linear and proportional relationship between Sr^{90} activity in soil in 1955 and the amount of rain for sites located in the Mediterranean area. It was pointed out that these sites are subject to the same seasonal distribution of rainfall (a marked winter maximum and extremely dry summers) and that rainfall over the whole area is derived from air masses of similar mixing history. From an extensive monthly rain collection network in the United States in 1956, Collins and Hallden[6] pointed out that good correlations could be attained within limited geographical areas. Walton[2] came to the same conclusion from a study of individual rain collections over a period of years at Westwood, New Jersey. In all these studies, it was indicated that the Sr^{90} concentration in rain, over a period of a month or more, was independent of the amount of rainfall.

In the case of an individual rain at a specific site, the Sr^{90} concentration has been shown to decrease as the storm progresses[2] and to vary with the height of the generating cloud.[7] In areas involving a number of different climatic regimes, it has been found that the Sr^{90}

concentrations are higher in regions of low precipitation than in areas of high precipitation within a given latitude.(2) In this regard, light rains are predicted to have higher Sr^{90} concentrations than heavier rains at a given station.

Although the importance of the contribution of dry deposition has been considered as a possible reason for calculated high Sr^{90} concentrations in low precipitation areas, there has been little concrete evidence for this from studies conducted so far.(1,2,8) Recent information from Norway suggests that at the end of June 1959, as much as 30% of the estimated Sr^{90} deposited may have been dry fallout.(9) Data from rainfall collections in the United Kingdom for the period 1957-1959 show that the cumulative fallout of Sr^{90} at 6 sites was proportional to the total rainfall at these sites.(4) The amount of Sr^{90} per unit volume of rain was approximately the same at each site and independent of the amount of rainfall over this prolonged period of time. The implication here is that dry deposition contributed insignificantly, if at all, at these United Kingdom sites and that all of the fallout during this period was brought down by precipitation.

As a supplement to the Atomic Energy Commission's world-wide soil sampling program, a study of the Sr^{90} deposition in a limited geographical area where the yearly amount of rainfall varies markedly from site to site, was conducted. In 1957, four sites in Clallam County, Washington on the Olympic Peninsula were selected where the mean annual amount of rainfall varies from 15 in. to 110 in. (Figs. 1 and 2). Four of the sampling sites are near the coast of the Juan de Fuca Strait and north of the Olympic National Park. A fifth site is further inland and just west of the National Park. The selection of these sites was based on two considerations: (1) the availability of an area suitable for soil sampling to obtain the total amount of Sr^{90} that had fallen out(10,11) and (2) the availability of reasonably accurate rainfall data from January 1953 to the date of sampling. January 1953 was selected as a starting period for the accumulation of rainfall data since only a very small amount of Sr^{90} had fallen out up to this time. The rainfall variation, proceeding westerly from Sequim to Forks, is extremely marked. The rainfall is lowest at Sequim, about 15 in. annually, and increases from one site to another in a westerly direction to Forks where it is about 110 in. annually. The sites were resampled in 1959 and 1960 with the addition of one more location, Clallam Bay. The annual rainfall for this period at each site has been very consistent and, in fact, has varied only slightly from long-term average figures published in 1939.(12)

The monthly rainfall pattern for the 5 sites is shown in Fig. 3. It is at once obvious that the seasonal trends are the same, with a low occurring in June, July and August and a high in December and January. It is interesting that the range is relatively small from site to site during the low rainfall period while the largest divergence in rainfall amount occurs during the months of highest precipitation. These 5 sampling sites are, in all probability, subjected to rainfall from the same westerly air masses that pass over the peninsula from the ocean.

The soils collected in Clallam County have been analyzed for Sr^{90} by the HCl extraction method(13) and results for the samplings in 1957 and 1959 have been reported previously(10). It was concluded from these data that the Sr^{90} rainwater concentration decreased with increasing amounts of rainfall up to about 62 in./year, after which there seemed to be little, if any, effect of rainfall amount. This phenomenon was explained on the basis of certain meteorological hypotheses; briefly, that precipitation that originates from air with less moisture can produce higher concentrations than air with greater moisture since more air is needed to make the same depth of rainfall. The assumption is inherent that arid regions obtain rain from the drier air and that raindrops falling in arid regions undergo more evaporation than in humid areas, thus enriching the Sr^{90} in the remaining liquid water. The contribution of dry fallout was not considered to be a primary reason for the decreasing Sr^{90} concentrations in rainwater with increasing rainfall amounts.

In Fig. 4, the measurements on 1960 samples of cumulative Sr^{90} activity per unit area are plotted against the amount of rainfall from 1953 through 1960. All available data for each sample, which include blind duplicate assays, re-analyses and blind fusions, are shown on the graph. The straight line is drawn through the averages for each site. The standard deviations from the means range between 8 and 14%. The linear relationship between Sr^{90} deposition and rainfall amount in the Clallam County area is clearly shown in Fig. 5 for the average results obtained for 1957 and 1959 soil collections as well as for the most recent samples collected in 1960. The dotted line portion of each graph is an extrapolation to zero rainfall on the assumption that the relationship between Sr^{90} activity and rainfall continues to be linear in this region. If this is indeed the case, then it would appear that there is a significant amount of Sr^{90} that has deposited on the soil in this area, which was not scavenged by rainfall. The cumulative fallout per inch of rain, as one would expect, increases from one year to the next as evidenced by the change in slope of the straight lines: 0.04 mc $Sr^{90}/mi.^2/in.$ in 1957, 0.10 in 1959, and 0.14 in 1960. The y intercept, which is assumed to represent the cumulative dry deposition, is 12 mc $Sr^{90}/mi.^2$ in 1957, 15 in 1959 and 16 in 1960. Fig. 6 is a plot of the activity of Sr^{90}/in. of precipitation (i.e., the Sr^{90} concentration in rainfall) as a function of the amount of precipitation. For each sampling year there is a definite decrease in the Sr^{90} concentration with increasing rainfall. It naturally follows that this would be the case if the straight lines intercept the y axis or, in other words, if there is a significant contribution of Sr^{90} which has fallen out during dry periods. If one makes the further assumption that the same amount of cumulative dry deposition has occurred at each site during each sampling year, (it will be recalled from Fig. 3 that during the coincident drier periods of the year, the rainfall difference from site to site is quite small) then by subtracting this amount from the total deposition, one obtains the contribution from rainfall alone. Then a plot of the activity of Sr^{90} versus rainfall produces a straight line which passes through the origin (Fig. 7) and the

plot of Sr^{90} concentration in rain versus the quantity of rain results in a horizontal straight line, indicating no correlation (Fig. 8). It would appear then, that Sr^{90} concentration in rain is, in fact, independent of amount of rainfall provided that one corrects for the Sr^{90} which has fallen out in the dry state, by gravitational settling or vertical impaction.

The period from the fall of 1957 through the fall of 1960 is of particular interest. This was the previous period of highest fallout rates due to intensive weapons testing over relatively short periods of time. In the Clallam County area it is estimated that 4 mc $Sr^{90}/mi.^2$ was deposited as dry fallout during this time as compared to 12 mc $Sr^{90}/mi.^2$ which fell out in the dry state up to 1957. A larger proportion of weapons detonated during the period 1957-60 were of high yield and high altitude as compared to those detonated prior to 1957. Debris originating from thermonuclear explosions will for the most part enter the stratosphere and then penetrate gradually into the higher troposphere where the probability is high that it will be scavenged to the earth's surface by rainfall. It is, therefore, expected that the ratio of fallout in precipitation to that collected in dry periods would be much higher during the period 1957 through 1960 than prior to this time. It is found, for the period 1957-60, that 0.29 mc $Sr^{90}/mi.^2/in.$ of rain or 4.4 $\mu\mu c$ Sr^{90}/l of rain fell out on the Clallam County area. The error associated with these figures is of the order of 20%.

The cumulative amount of Sr^{90} contributed as dry fallout in Clallam County is estimated to be 16 $mc/mi.^2$ up to the fall of 1960. The assumption has been made that this contribution is the same at each of the 5 sites irrespective of the amount of rainfall. This is thought to be reasonable because, as was previously pointed out and can be seen from Fig. 3, during the period of lowest rainfall there is only a small difference in amount of monthly rainfall among the sites. It is possible that during this period of time, coincident dry periods are not uncommon. If this is indeed the case, it is interesting to calculate the percent contribution from dry deposition of the cumulative measurements made in 1960 at each of the sites.

TABLE 1

Contribution to Cumulative Sr^{90} from Dry Deposition in 1960

Site	Sequim	Port Angeles	Joyce	Clallam Bay	Forks
% Contribution from Dry Deposition of Sr^{90}:	47	35	21	15	12

In summary, the Clallam County study has shown that there is a remarkably good linear relationship between Sr^{90} fallout and rainfall amount. If it is assumed that all of the Sr^{90} measured was brought down by rain,

then the Sr^{90} activity per unit volume of rain decreases with increasing rainfall. Strong evidence is available, however, to infer that a substantial amount of the Sr^{90} deposited in Clallam County was not associated with precipitation. When the amount of Sr^{90} brought down by rainfall alone is calculated by subtracting the dry fallout contribution from the total measured, then the Sr^{90} concentration in rain is seen to be independent of the amount of rainfall.

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TABLE 2

Rainfall and Sr^{90} Data for Callam County Soil Sites

	In. of precipitation from 1-1-53 thru date of collection			mc $Sr^{90}/mi.^2$		
Site	1957	1959	1960	1957	1959	1960
Sequim	87	119	135	13.3	29.7	34.1
Port Angeles	134	187	215	20.6	36.2	46.4
Joyce	288	404	459	21.5	46.9	74.9
Clallam Bay	--	566	648	--	71.2	105
Forks	576	812	922	32.4	99.6	137

Figure 1

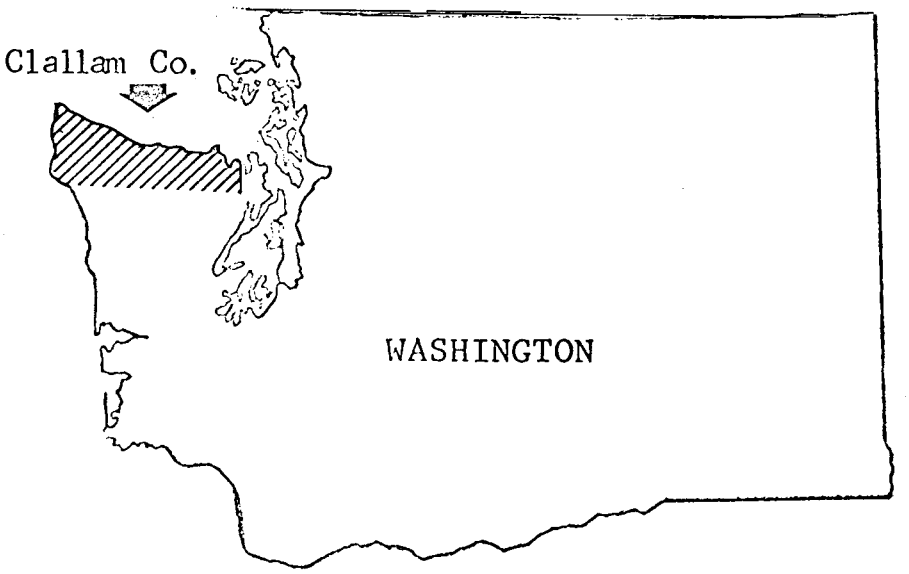


Figure 2

Clallam County, Washington, Olympic Peninsula:

Soil Sampling Sites for Measurement of Strontium-90

Deposition

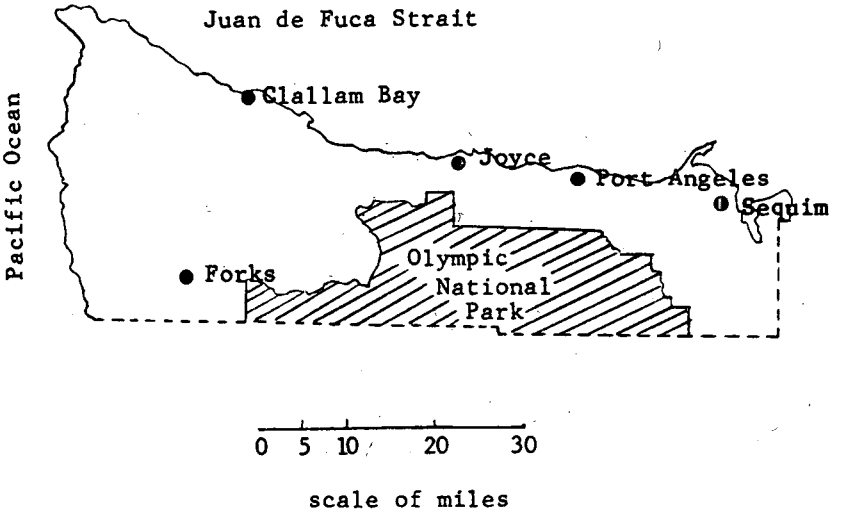


Figure 3

Mean Monthly Precipitation

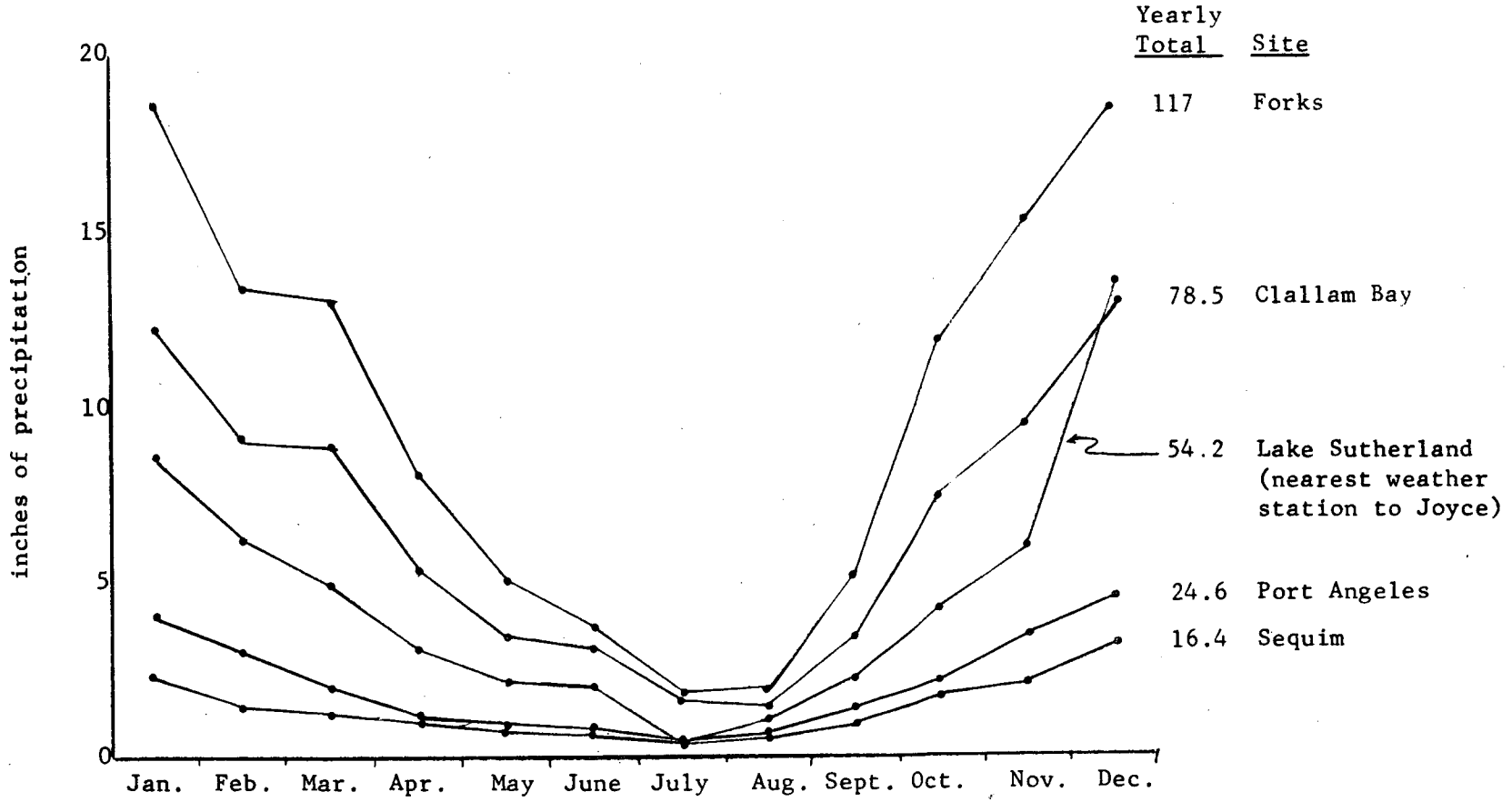


Figure 4

1960 Strontium-90 Deposition

as a function of

Amount of Precipitation

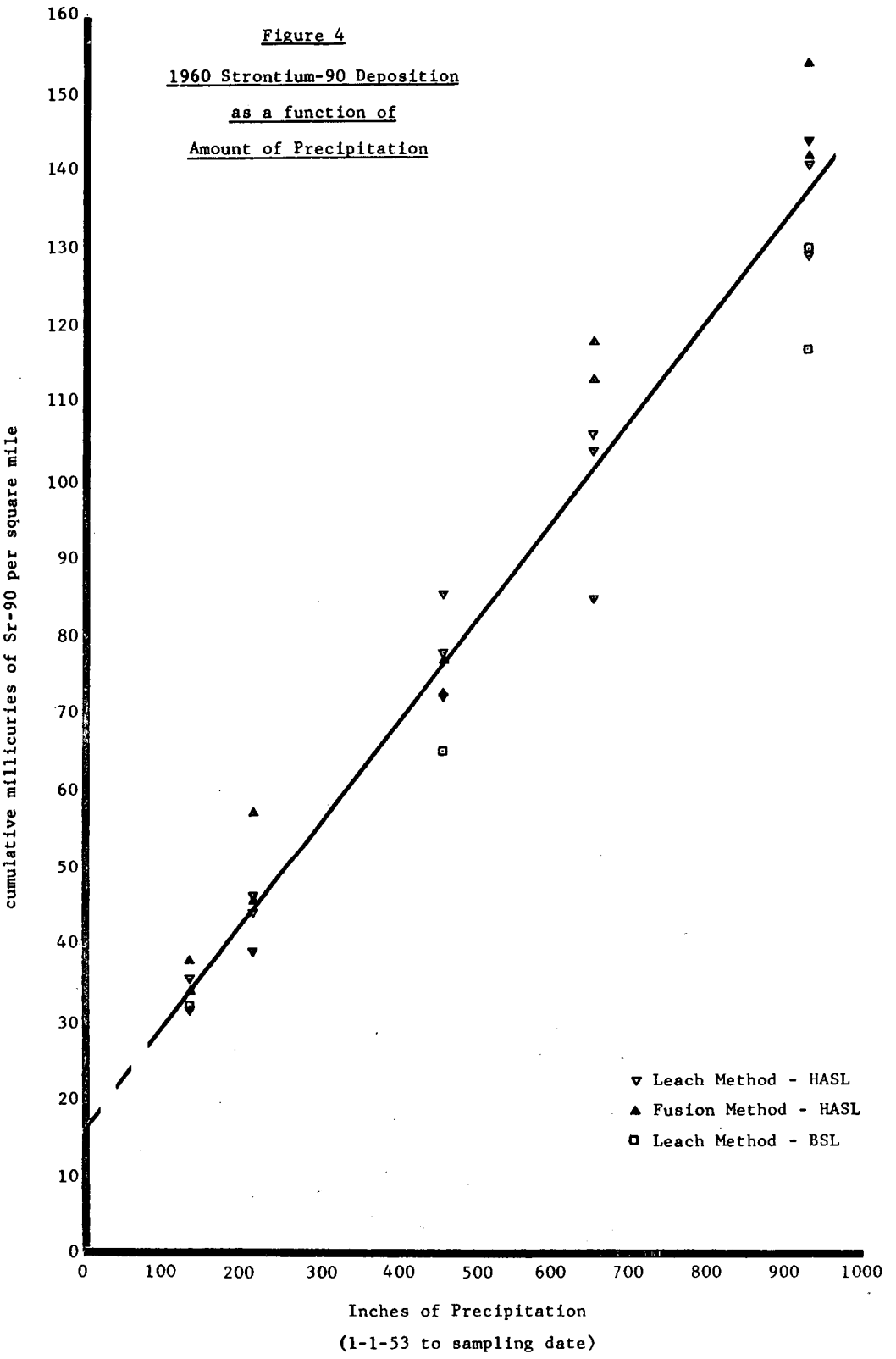


Figure 5

Strontium-90 Deposition as a Function of Amount of Precipitation 1957, 1959 and 1960

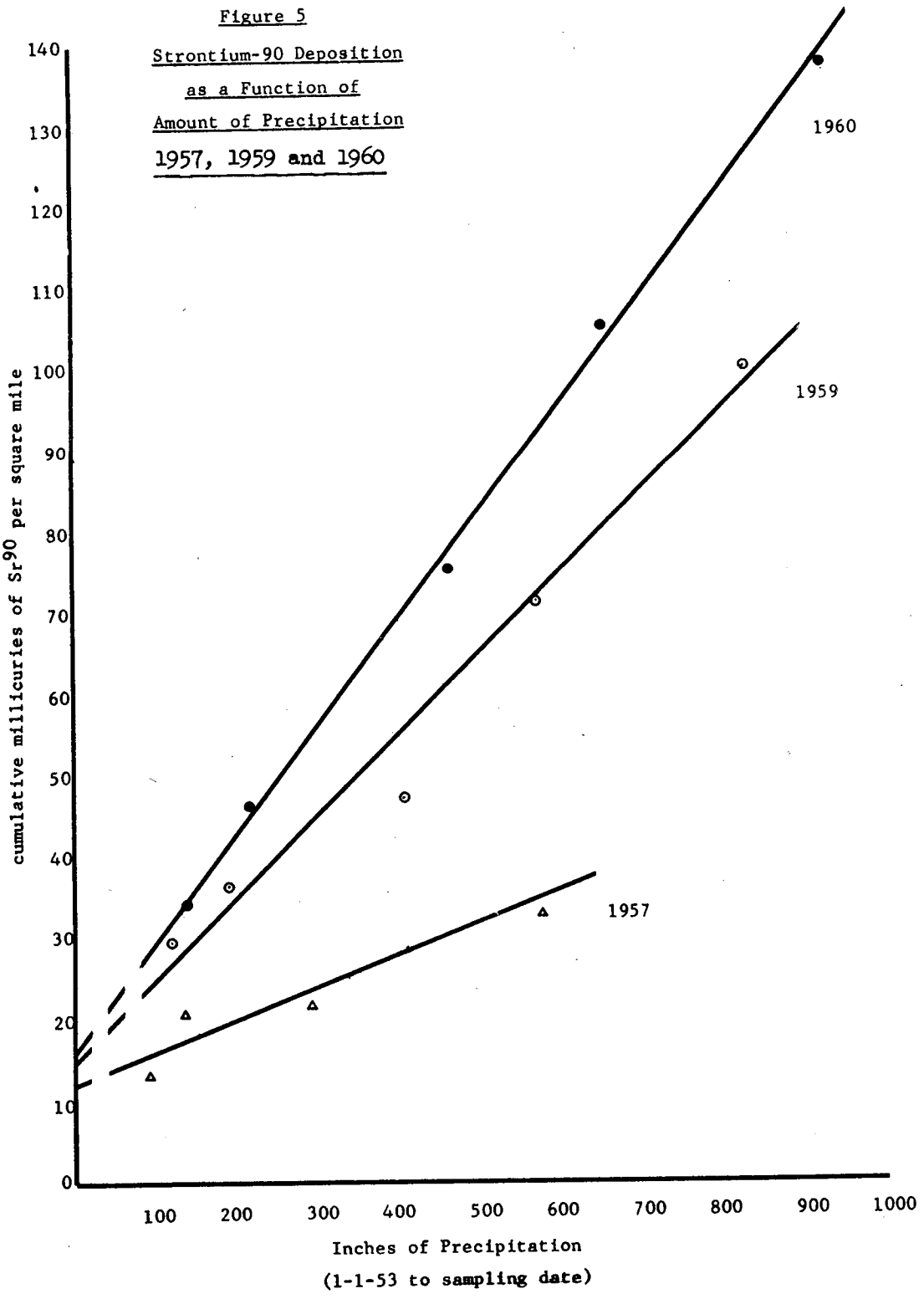


Figure 6

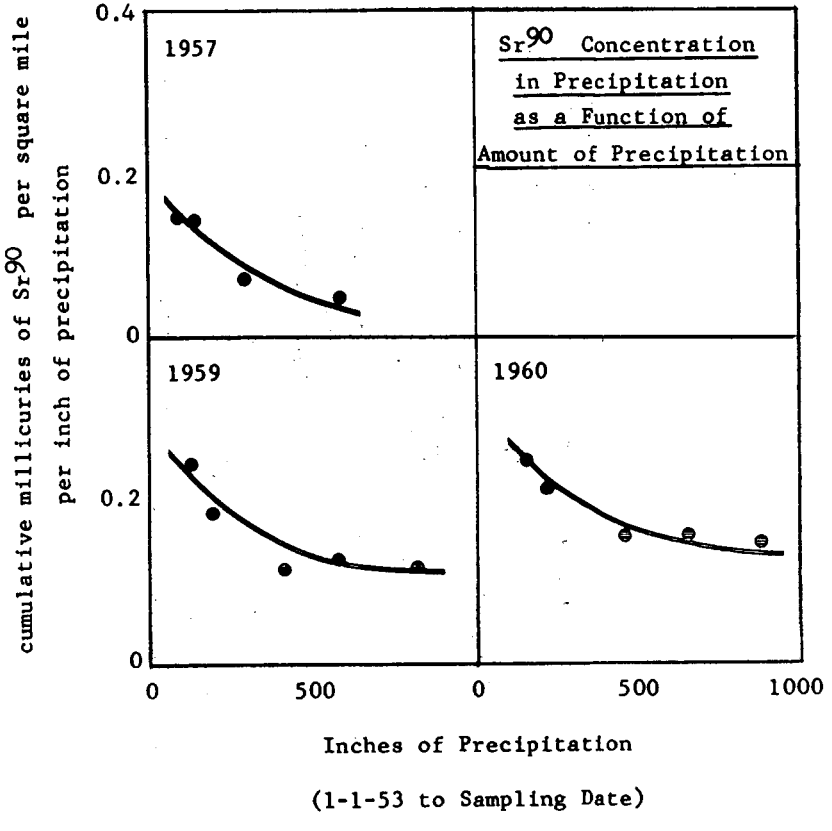


Figure 7

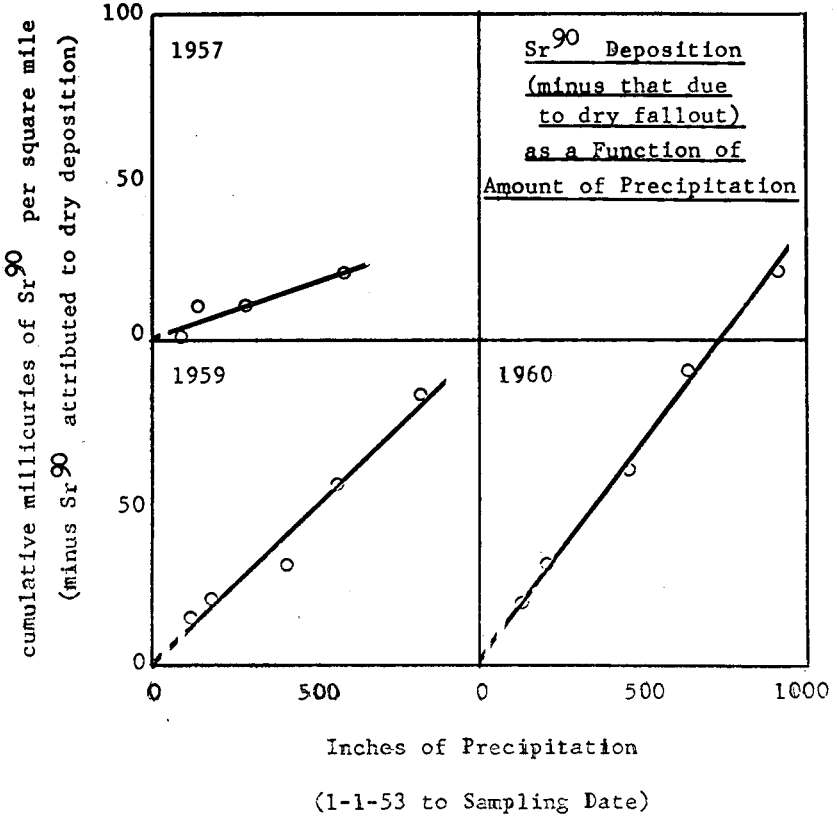
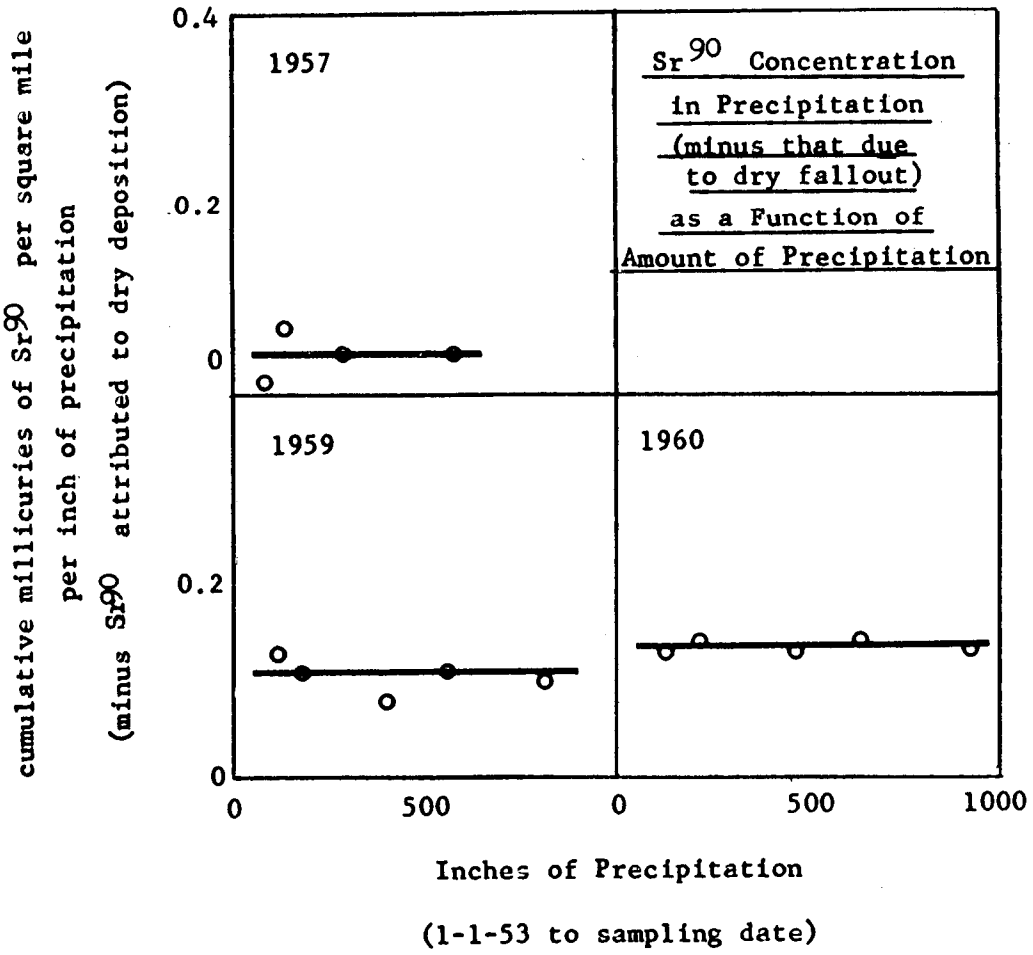


Figure 8


cumulative millicuries of Sr90 per square mile
per inch of precipitation
(minus Sr90 attributed to dry deposition)
0.4
0.2
0
0.2
0
1957
Sr90 Concentration
in Precipitation
(minus that due
to dry fallout)
as a Function of
Amount of Precipitation
1959
1960
0
500
0
500
1000
Inches of Precipitation
(1-1-53 to sampling date)

RUNOFF OF STRONTIUM-90 FROM AGRICULTURAL LAND SURFACES

Maurice H. Frere and Ronald G. Menzel
U. S. Soils Laboratory
Soil and Water Conservation Research Division
Agricultural Research Service
U. S. Department of Agriculture
Beltsville, Maryland

Sr^{90} is deposited on soil surfaces chiefly through rainfall (4). When surface runoff and soil erosion occurs, some movement of recently deposited Sr^{90} would be expected. The extent of such movement has been studied by measurements on the soil in sloping areas, on runoff from small plots, and on river waters. The number of measurements are quite limited and serve only to outline the general features of Sr^{90} movement in runoff.

Runoff From Small Watersheds

Sr^{90} contents were determined in soil samples taken in June 1960 from small watersheds near Coshocton, Ohio. The watersheds were on predominantly Muskingum and Keene silt loams with slopes averaging from 9 to 16%. The area of the watersheds was usually from 1 to 2 acres. The watersheds are maintained by the U. S. Hydrological Research Station, and records of soil erosion, water runoff, rainfall, and soil management practices have been kept since before the advent of nuclear weapons testing.

Two watersheds in permanent pasture with little water runoff were selected as reference points. The Sr^{90} content of the surface soil to a depth of 6 1/2 in. was 160 pc/kg. The bulk density of this soil is approximately 1.4 g/cm^3 (2). Thus, the calculated Sr^{90} content is approximately 90 $mc/mi.^2$ This may be compared with an estimate of

75 mc/mi.^{2} obtained from the content in soil as of late 1959 (1) and in precipitation during the following winter and spring (3). It appears that little or no Sr^{90} had been lost from the watersheds in permanent pasture.

Six cultivated watersheds (two each in corn, wheat, or meadow) were sampled along several contours and at the entrance to the spillway at the base of the watershed. These watersheds had been maintained in a 4-year rotation of corn-wheat-meadow-meadow since the beginning of Sr^{90} fallout. The Sr^{90} content of the surface soil to a depth of 6 1/2 in., which included the entire plow layer, averaged from 65 to 100 pc/kg (Table 1). From other measurements (2), the bulk density of the cultivated watersheds might be expected to range from 1.2 to 1.35 g/cm.^{3} Thus, the plow layer in cultivated watersheds had lost from 1/3 to 2/3 of the Sr^{90} on it.

TABLE 1

Average Sr^{90} content and water runoff on small watersheds at Coshocton, Ohio, as related to cropping history and slope.

Crop			Average Slope	Sr^{90} (pc/kg)	Water Runoff (in.)		
1958	1959	1960	(%)	June, 1960	1958	1959	Jan.-June 1960
--	Pasture	--	20	153	0	0	0
--	Pasture	--	20	163	0	0	0
Meadow	Meadow	Corn	16	64	1.32	1.88	1.77
Meadow	Meadow	Corn	15	66	0.21	1.41	0.34
Meadow	Corn	Wheat	9	73	1.22	3.91	0.58
Meadow	Corn	Wheat	11	79	0.71	2.02	0.35
Wheat	Meadow	Meadow	13	100	0.18	1.36	0.31
Corn	Wheat	Meadow	16	78	1.17	3.57	0.89

Most of the Sr^{90} lost from these plots was probably carried in surface runoff. On pairs of plots with similar cropping histories, it appears that more Sr^{90} had been lost from the plot with greater water runoff during the last 2 1/2 yrs. prior to sampling. During this period the major portion of Sr^{90} fallout occurred (3). However, the differences

in Sr^{90} content are greater between plots with different cropping histories, and on these the relationship to water runoff is not clear. The concentration of Sr^{90} in water runoff probably depends on crop cover, length and steepness of slope and soil type in addition to the concentrations of Sr^{90} on the soil surface and in the curren rainfall.

Other factors which may have caused appreciable loss of Sr^{90} from the cultivated watersheds are crop removal and leaching into the soil. The amount lost by crop removal probably did not exceed 5% of the Sr^{90} fallout. That lost by leaching probably did not exceed 15% of the accumulated Sr^{90} fallout at the time of sampling.

Somewhat lower Sr^{90} contents were found, on the average, in soil samples from lower contours on the watersheds (Table 2). This indicates some effect of length of slope on removal of Sr^{90} in water runoff. There was little, if any, accumulation of Sr^{90} in the area of silt deposition at the entrance to the spillway. The Sr^{90} lost must have been carried completely off the watersheds.

TABLE 2

Average Sr^{90} content on different contours of six small, cultivated watersheds, Coshocton, Ohio, June 1960.

Watershed Contour	Sample Depth (in.)	Sr^{90} (pc/kg of soil)
Top	0-6 1/2	77
Middle	0-6 1/2	75
Bottom	0-6 1/2	68
Entrance to spillway	0-8	86

Runoff From Small Plots

In an earlier study (5), the transport of Sr^{90} in runoff from small plots was shown to be affected by vegetative cover and to be related to the amount of soil eroded from the plots. One set of plots was located at La Crosse, Wisconsin, on Fayette silt loam with a 16% slope. Another set was located at Tifton, Georgia, on Tifton loamy sand with a 3% slope. The length of the plots was 72.5 ft. at La Crosse and 83 ft. at Tifton. The losses of Sr^{90}, soil, and water in runoff from these plots are summarized in Tables 3 and 4.

TABLE 3

Transport of Sr^{90}, soil, and water in runoff from plots in corn, oats, and clover at La Crosse, Wisconsin (March 13-August 18, 1957).

Crop	Sr^{90}* (pc/ft.^{2})	Amount in Runoff from Plot	
		Soil (tons/acre)	Water** (in.)
Corn	11.5	1.11	0.93
Oats	10.5	1.22	1.25
Clover	0.9	0.03	0.15

* Total fallout of Sr^{90} during this period was 257 pc/ft^2.

** Total precipitation during this period was 22.55 in.

TABLE 4

Transport of Sr^{90}, soil, and water in runoff from plots in corn, oats, and peanuts at Tifton, Georgia (March 11-December 2, 1957).

Crop	Sr^{90}* (pc/ft.^{2})	Amount in Runoff from Plot	
		Soil (tons/acre)	Water** (in.)
Corn	2.5	0.70	1.32
Oats	0.8	0.34	0.37
Peanuts	2.6	0.82	1.20

* Total fallout of Sr^{90} during this period was 180 pc/ft^2.

** Total precipitation during this period was 40.91 in.

The plots with the least exposure of bare soil during the period of the study had the lowest losses of Sr^{90}, soil, and water. At La Crosse, the soil was bare at the time of seeding oats or corn (In April or May, respectively) and several weeks were required to establish good ground cover. The crop on the clover plot had been established the preceding year and it provided good ground cover during the entire period of the

study. At Tifton, the soil was bare at the time of seeding corn or peanuts (also in April or May, respectively). However, oats had been established the preceding fall at Tifton, and this crop provided good ground cover from the beginning of the period of the study until the stubble was disk harrowed in October.

Although the relation between transport of Sr^{90} and that of soil was previously noted (5), there appears to be a closer correllation between transport of Sr^{90} and inches of water runoff. At La Crosse, 1 in. of water runoff transported about 10 pc $Sr^{90}/ft.^2$ At Tifton, the same amount of runoff transported about 2 pc $Sr^{90}/ft.^2$ Crop cover seemed to have little effect on the amount of Sr^{90} transported per inch of water runoff.

It should also be pointed out that the concentration of Sr^{90} in the precipitation at La Crosse was 11 $pc/ft.^2/in.$ of precipitation, or nearly the same concentration that was observed in the runoff. At Tifton, the concentration was 4.4$pc/ft^2/in.$ of precipitation, or about double the concentration that was observed in the runoff. The greater retention of Sr^{90} from precipitation at Tifton than at La Crosse is probably related to the lower slope or sandier type of soil.

The average runoff of Sr^{90} at La Cross amounted to about 3% of the fall-out, and at Tifton it amounted to about 1%. At Coshocton, the average runoff of Sr^{90} was much higher, perhaps from 20 to 40%. The average water runoff at Coshocton for the period from 1954 to 1960 was about 5% of the precipitation. Thus, it appears that the concentration of Sr^{90} in runoff at Coshocton was higher than at either La Crosse or Tifton.

Runoff in Rivers

If little of the Sr^{90} in runoff is desposted at the base of slopes, as at the Coshocton watersheds, then it must appear in streams and rivers. Some could be deposited in impounded waters, along stream beds or it could be carried into the oceans. Numerous analyses of river waters have been made, and calculations have indicated that, during the first quarter of 1959, from 4 to 12% of the Sr^{90} in precipitation in the Ohio River basin appeared in the river water (6).

Enough data are now available so that similar calculations can be made for other river basins and other time period. The concentration of Sr^{90} in the river water has been determined quarterly on composites of weekly samples (7). This concentration may be multiplied by the total water flow during the quarter (8) to determine approximately the amount of Sr^{90} carried by the river. If this amount is divided by the drainage area at the sampling point, one may obtain a value for runoff in mc/mi^2. The estimated deposition of Sr^{90} in precipitation within the drainage area may be derived from data for U. S. AEC Health and Safety

Laboratory collection stations within the area (3). Examples comparing the amounts of Sr^{90} in precipitation and runoff are shown in Tables 5 and 6.

TABLE 5

Sr^{90} in precipitation and runoff in the Ohio River basin above Evansville, Indiana, from October 1, 1958 to September 30, 1959

Quarter	Sr^{90} Content Precipitation* (mc/mi^2.)	Runoff (mc/mi^2.)	% of Sr^{90} in Runoff
1958 IV	1.74	0.08	4.6
1959 I	6.45	0.86	13.3
1959 II	8.12	0.43	5.3
1959 III	1.03	0.14	13.6

*Sr^{90} deposited in precipitation at Pittsburgh, Pa., and Louisville Ky., were averaged to estimate the deposition for the entire area.

TABLE 6

Sr^{90} in precipitation and runoff in the Missouri River basin above Yankton, South Dakota, from April 1, 1959 to September 30, 1960

Quarter	Sr^{90} Content Precipitation* (mc/mi^2.)	Runoff (mc/mi^2.)	% of Sr^{90} in Runoff
1959 II	4.94	0.036	0.7
1959 III	0.96	0.009	0.9
1959 IV	0.25	0.006	2.3
1960 I	0.39	0.013	3.3
1960 II	0.88	0.014	1.6
1960 III	0.48	0.017	3.5

* Sr^{90} deposited in preceipitation at Helena, Montana, and Williston, North Dakota, were averaged to estimate the deposition for the entire area.

The percentages of Sr^{90} appearing in the river waters are consistent with the observed losses from watersheds and small plots. The percentages in river waters are averages for a variety of land surfaces. It is to be expected that within each drainage area some land surfaces will have essentially no runoff of Sr^{90} and others will have a percentage of runoff several times higher than that observed in the river water.

Present data are inadequate to show variations in the amount of Sr^{90} runoff from various land surfaces. Although the analysis of river waters gives an estimate of the amount of Sr^{90} removed from large areas, the amount of Sr^{90} deposited in impounded waters or along stream beds is still unknown.

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THE POSSIBILITY OF GROUND WATER CONTAMINATION BY FALLOUT*

Alfred Clebsch, Jr.
U. S. Geological Survey
Washington, D. C.

and

Joseph A. Lieberman**
Division of Reactor Development
U. S. Atomic Energy Commission
Washington, D. C.

In considering water-supply problems that could arise from contamination of streams and reservoirs by bomb debris in the event of a nuclear attack, the idea has been generally accepted that water supplies derived from ground-water sources can be expected to be free of contamination. As with most generalizations, there are possible exceptions.

It is the purpose of these remarks to describe briefly the kinds of geohydrologic terranes that by reason of their geologic structure and mineralogic characteristics provide the highest degrees of safety from fallout contamination, and also to indicate the type of ground-water reservoirs that are susceptible to contamination as a result of massive deposition of bomb debris.

Research on the disposal of radioactive wastes into earth materials has made substantial contributions to our understanding of the role of mineral reactions in the uptake of radionuclides from solution, but much more work is necessary before the knowledge gained by this research can be extended in a quantitative way to terranes other than those in which the major nuclear activities are located.

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*Publication authorized by Director, U. S. Geological Survey.
**Assistant Director for Nuclear Safety.

With the notable exception of tritium, there appears to be almost no indication of the detection of fallout in ground water up to the present time. Certainly there has been no indication of concentrations of fallout-produced radioactivity of health significance in ground water. This fact alone tends to bear out the idea that ground-water reservoirs would be relatively free from contamination during and after a nuclear attack. It might also be reasoned that since so much fallout consists of particulate matter, its apparent absence from ground water might be explained solely on the basis of removal by sedimentation or natural filtration. These factors are extremely important, but a more complete explanation is provided by a consideration of the mechanisms of ground-water accumulation and movement.

Virtually all ground water is part of the hydrologic cycle; that is, it originates as precipitation which infiltrates below the root zone of plants, and moves into subsurface reservoirs. Under the influence of pressure gradients that develop as a consequence of gravity and differences in permeability of the soil and rock, ground water moves laterally from areas of replenishment, or recharge, toward springs or surface streams, from which it may flow over the surface to the sea, or be discharged directly back into the atmosphere by evaporation or transpiration.

Movement of water within a ground-water reservoir is extremely slow. Speaking of an average rate of ground-water movement is not very meaningful when one considers that velocities through core samples of rock and soil have been measured in the laboratory over a range of 8 or 9 orders of magnitude. However, in most aquifers the rates of movement are on the order of feet per day. In highly permeable aquifers rates of several hundred feet per day have been measured. The rates of movement are infinitesimal when compared to flow rates in surface streams. This factor provides a considerable holdup time in most ground-water reservoirs during which decay of radionuclides can take place. Other physical and chemical processes, such as sorption and dispersion, tend to reduce the concentration of fallout materials in solution, but not to the same degree for all ions nor in all types of geologic materials.

Hydrologic and geologic characteristics that favor rapid infiltration, relatively rapid movement, and relatively little contact with the rock skeleton of the aquifer indicate types of ground-water reservoirs which, in the event of heavy fallout, could become contaminated.

These conditions are approached in near-surface reservoirs composed of cavernous limestone, basalt (also cavernous in places), and highly permeable granular materials, such as water-laid deposits which consist dominantly of well-rounded, evenly-sized fragments. In even these highly permeable geohydrologic terranes ground water is

replenished by the fraction of precipitation or runoff that must seep through soil, or the beds and banks of streams; thus ground-water recharge is nearly always subject to some filtering action. However, in limestone, basalt, and "permeable-alluvial" terranes, the filtering action is at a minimum, and rates of movement are highest. In some limestone aquifers the filtering action is so poor that well-water becomes turbid after heavy rains.

In the United States, limestone aquifers that are highly susceptible to fallout are important in the south and east, particularly in Florida, in the interior lowlands of the Ohio and Mississippi Valleys, and on the Ozark plateau.

These areas are predominantly in the humid parts of the country, where the water table is seldom more than a few feet or a few tens of feet below the surface. Further, in limestone the pore space through which ground water moves is commonly of secondary origin; that is, the openings were formed after the rock was deposited and were subsequently enlarged by the solution action of circulating ground water. In openings of this type, there is rapid flow and a minimum amount of contact between the ground water and its enclosing rock.

Basalt aquifers are of widespread importance only in the western United States. The best known examples are those of the Snake River plain in southern Idaho, and the Columbia River basalts of Washington and Oregon. Elsewhere in the West basalt aquifers are rather limited in areal extent and are of only local importance.

Because the climate in so much of the West is semiarid to arid, the ground-water reservoirs in basaltic terranes are commonly deep below the surface. Depths to water are measured in hundreds of feet or many tens of feet. In many places a considerable thickness of granular material--sand, gravel, silt, or clay--occupies the space between the land surface and the water table and recharge to the ground-water body must move through these deposits before reaching the basalt aquifer. Thus, an opportunity is provided for filtration of particulate matter and sorption of radioactive ions.

The void spaces in a basalt aquifer are highly varied in size, shape, and distribution. On the one extreme they are cavernous tubes, formed by the characteristics of the lava flows. Water flows through these openings just as through pipes. On the other extreme are interflow layers that are filled with angular blocks, cinders, and volcanic ash. Obviously, these layers offer much more resistance to flow and greater opportunity for filtration and sorption than do the open conduits.

Permeable granular aquifer materials are well down the scale of susceptibility to contamination by fallout as a general rule, but in certain geohydrologic situations possibilities of contamination should

be considered. An aquifer in the alluvial deposits beneath the flood plain of a river, which is recharged by the river, is a typical example. Shallow aquifers in areas of very sandy soil are susceptible also. These situations are usually of limited areal extent, but occur quite commonly in many parts of the country.

Aquifers that could be expected to yield the most dependable contaminant-free sources of water (after a nuclear attack) are characterized by great depth to water in the recharge area, granular nature, high sorptive capacity, and clay beds between the aquifer and the land surface in the areas of ground-water utilization. Deep burial provides a thick section of earth materials through which surface water must infiltrate to reach the aquifer, and hence, a good opportunity for uptake of contaminants in the zone of aeration above the aquifer. Granular materials provide additional protection because of the lower ground-water velocities found in them, as well as much greater surface areas for mineral reactions than in non-granular aquifers. Artesian aquifers, in which the water-bearing units are confined below rock units sufficiently impermeable to inhibit upward flow and to maintain hydrostatic pressures are most favorable, because the confining layer--usually clay or shale--will also inhibit or effectively prevent the flow of contaminants downward into the aquifer. Sandstone aquifers of the Atlantic and Gulf Coastal Plains are typical of this group, although artesian aquifers occur in many parts of the country.

Tritium is of particular interest in any discussion of ground water and fallout, because of the occurrence of bomb-produced tritium found in certain aquifers in recent years. Since the pioneering work of W. F. Libby and other investigators at the University of Chicago, there has been a growing interest among hydrologists in the use of tritiated water as a tracer. The atmospheric tritium peak produced in 1954 during the Operation CASTLE series of weapons tests has been used as a reference point in time for a number of ground-water investigations. The event has provided a basis for estimates of the rates of ground-water movement, as well as for studies of depths and rates of mixing and other hydrologic considerations.

Investigations of this type are particularly important for the knowledge they provide on holdup time and the amount of dilution to be expected in an aquifer--factors that are of particular interest in assessment of possible contamination by radionuclides. Atmospheric tritium peaks that (presumably) have resulted from recent Russian tests will provide an additional time marker for future hydrologic studies. The unique value of tritium as a tracer stems from the fact that it is traced as water, thus avoiding some of the difficulties that arise in the use of other tracers, such as density differences and adsorption.

The rates of movement of all cationic solutes will lag behind that of tritiated water because of the sorptive effects of the aquifer material.

Tritium monitoring of ground water can therefore provide advance notice of potential contamination from other, more hazardous nuclides. It is important to note here, however, that comprehensive studies of aquifers that are susceptible to contamination would be necessary to develop quantitative relations between the travel time of tritium peaks and the probable breakthrough times for unacceptable concentrations of other radionuclides.

As part of civil defense planning, studies should be made of emergency sources of water, not only from the standpoint of treatment plants, pumping and distribution facilities, and other utilities, but also of the geologic and hydrologic aspects of the supply.

An inventory of wells, including information on their depth, yield, general condition, pumping plants, geologic unit from which the water is obtained, and other important facts, should be part of any detailed civil defense plan.

The fixation or retention of nuclides in soil and in aquifer materials is primarily a function of the interactions between ions in water solution and mineral particles. Extremely complex chemical and hydrodynamic systems are involved, and although a large body of knowledge has been accummulated in the fields of soil chemistry and physics, clay mineralogy, and crystal chemistry, many new problems have arisen, partly from the fact that the concentration of radioactive ions is so small in relation to the concentrations of other ions in solution. New knowledge has also been required in the field of hydrodynamics, particularly on the effects of inhomogeneities of earth materials on permeability and, hence, on the dispersion of dissolved ions. Contributions to the solution of the first problem have been made by several workers in the field of waste-disposal research. Basic studies of the role of clay minerals in removing radioactive ions from solution have been made by T. Tamura and D. G. Jacobs at Oak Ridge, H. C. Thomas at the University of North Carolina, J. L. Nelson and others at Hanford, C. R. Naeser and others of the U. S. Geological Survey, and by W. R. Kaufman at the University of California.

Tamura and Jacobs have studied kaolinite, montmorillonite, illite, and vermiculite from the standpoint of the mutual effects of crystal structure and fixation of cesium and strontium. Thomas has studied the kinetics of mineral reactions with radionuclides from the standpoint of chemical thermodynamics. At Hanford the work has involved equilibrium studies in which soil-water suspensions were investigated with reference to nuclide concentration, pH, equilibrium time, and species and concentration of complementary ions. Later work has

included additional variables and the use of soil columns. Other studies at Hanford have included the role of carbonate and phosphate minerals as well as natural zeolites in retention of radionuclides. The Geological Survey group has investigated variations in the sorptive capacity of vermiculite, glauconite, crandallite, and anhydrite with changes in pH. The laboratory work under Kaufman at the University of California has included both the chemical kinetics and hydrodynamic aspects of the problem; it has been followed by field experiments using labelled water. Reference clay minerals and samples of a typical oil sand have been investigated, with particular reference to the effect of calcium concentration on cesium exchange. The studies of hydrodynamics have been aimed at understanding principles involved in the injection of liquid wastes into permeable formations, but valuable information on ground-water flow has resulted from the work.

Other work on hydrodynamics has been carried out at Idaho and at Hanford and by C. V. Theis and H. E. Skibitzke of the Geological Survey. The Idaho work has been concerned with movement in complex lava formations. Work at Hanford has concentrated on unsaturated flow above the water table where computer solutions of the complex mathematical relationships have been necessary. The Geological Survey efforts have been aimed at explaining dispersion in terms of spatial variations in permeability that result from inhomogeneities in aquifer materials.

In general all of this work has provided a better insight into the physical and chemical interactions between radionuclides, earth materials, and water.

In closing, passing mention should be made of investigations related to the behavior of radioactive materials in surface streams and flow of radioactive material into surface reservoirs. Notable among the former are studies in the Clinch River below Oak Ridge by Oak Ridge National Laboratory, U. S. Geological Survey, U. S. Public Health Service, Tennessee Valley Authority and several State agencies. For the latter, reference is made to the work of several years ago by Thomas and co-workers in the Framingham, Massachusetts, reservoir and by Kilcawley in the Troy, New York, reservoir. In both cases the "coefficient of radioactive runoff" was quite low, that is, concentration in the reservoir could almost be accounted for by direct fallout on the water surface.

DISCUSSION

Mr. Alfred W. Klement, Jr., Chairman of the session, directed the discussion of the three review papers presented.

KLEMENT: Dr. ALEXANDER, you didn't take all of your time. Did you have any other comment you wish to make now? I am not sure whether you had already finished or were trying to save some time for us.

ALEXANDER: No, I had really finished. I might remark, that after listening to all of the talk about height of cloud and first part of the rain and last part of the rain, it seems that time and integration is a wonderful smoother of points on a curve.

Lloyd R. SETTER* (U. S. Public Health Service, Cincinnati, Ohio): I was just wondering what were the levels of tritium with which you were concerned.

CLEBSCH: These levels are expressed in tritium units, which is defined as the ratio of tritium atoms to 10^{18} protium (H^1) atoms. This is a field on which I am far from expert, but there is quite a bit of literature on the subject. Tritium is produced naturally by cosmic radiation, but the most easily traceable source is bomb tritium. In ground water, numbers of the order of 50 to 150 tritium units are not uncommon.

LIST: I have a comment on Dr. ALEXANDER's paper. I think it should be pointed out that this is a very homogeneous set of rainfall data. It is mainly orthographic rainfall. And the conclusions may not necessarily be valid for other types of rainfall. However, I do think that his value of 15 mc $Sr^{90}/mi.^2$ of dry fallout is a very significant finding and if this is true (and it is true over most of the U. S.), then we can say that somewhere between 15 and 20% of the fallout we observe in the eastern part of the United States is dry fallout.

* It has been suggested that the question was asked by POLSTER.

ALEXANDER: I don't think that requires any comment from me. Mr. LIST knows as much about this as I do. I agree with his statement. This is consistent with what we find for dry fallout in the Imperial Valley, and at Antofagasta in Chile there was less than 1 mc $Sr^{90}/mi.^2$ in 1960. The Southern Hemisphere, apparently, at least at the latitude of Antofagasta, didn't get much dry fallout.

QUESTION: I think the question is whether, if this dry deposition is going to have any effect on the inventory of the budgeted fallout, that would make the total deposition higher or lower.

LIST: Since any of these calculations are based essentially on observed deposition, if the fractional relationships remain constant, I imagine our estimates will remain constant. That is, they won't affect the estimates particularly.

QUESTION: When Dr. MACHTA (?) said, "Treated for rainfall in a proper way", what does that mean?

LIST: It is based on observations that were made following the 1958 test series. The soil data also includes the dry fallout. So I don't think it would make any serious difference.

Raymond R. EDWARDS (Nuclear Science and Engineering Corp., Pittsburgh, Pa.): Since the question of units has been raised, it seems to me this might be a good time to begin trying to clear up this problem which is confusing everybody in the field. The International Atomic Energy Agency has adopted standard units. The "micromicrocurie" is much more difficult to work with than the "picocurie" in writing things out, and this has been adopted in their reports, and I don't know why it shouldn't be adopted throughout and applied to tritium as well as to natural radioactive elements and fallout elements. The argument is that there is too much literature already in terms of other units. This is true, but this can be turned around and used as an argument for standardizing as soon as possible, which has happened in other fields.

KLEMENT: I certainly agree on the standardization of units. However, I suspect that unless we pass a law where it is a criminal offense not to use such units, we are not liable to have much standardization.

HOLLAND: I feel impelled to elaborate a little on what Bob LIST said. This question of the curvilinear relation between the concentraion of Sr^{90} entering rain, and the amount of rain, has interested me for some time. I think there are really three different kinds of cases. The one that Dr. ALEXANDER treated is one in which you have essentially a homogeneous climate except for the amount of precipitation. There is another one which you get by looking at the variation over a wide range of climates, but, say, a simulated situation in which the concentration

of radioactivity in the air averaged over a long time might be considered homogeneous. So, you have the effect of climate, that is, the type of air masses out of which the precipitation falls and the average humidity conditions of the area. And I think that that curvilinear relation is one which might not disappear under the type of treatment that Dr. ALEXANDER used. The third one is what you might call the microscopic case where you can look at the concentration as a function of total precipitation in individual rainstorms. You look at small rainstorms or small amounts of deposited precipitation, and large amounts, and you get a curvilinear relation there. This might also be a result of different mechanisms which occur during the actual precipitation formation process and evaporation on descent, and so on. I think what Dr. ALEXANDER has done represents a considerable headway in solving this problem by isolating one of these types and getting rid of it. I think there are still the others to be investigated.

Bentley GLASS (Johns Hopkins Univ., Baltimore; Advisory Committee for Biol. & Med., U. S. AEC): Two of the speakers have referred to the very slow removal of the radionuclides in the soil by runoff, and the last comment that was made was that in reservoirs one could account for the contamination largely on the basis of what falls on the water surface. I would like to hear something more said about the rate at which the settling out process would occur in standing reservoir waters, whether it remains fairly evenly distributed in the water or settles to the bottom relatively rapidly, so that we could have some idea about the duration of the contamination of reservoir water.

CLEBSCH: I cannot answer that question. Perhaps someone in the audience can.

KLEMENT: Would someone like to comment on this? (None was offered).

CLEBSCH: Perhaps this indicates a field that needs more study.

HOLLAND: I would like to see someone else comment, but I believe that certainly a contribution to the answer to this question would come from the studies of rain water and the solubility of the different nuclides in the collected water in fallout pots. Soluble and insoluble fractions have been measured, and this certainly would be a clue to how the fallout would at least initially appear, as between the water and the sediment. Then, of course, there is a question of what happens over a period of time while it is in the reservoir. Some of our radiochemists have done quite a bit of work on this.

ALEXANDER: You have another matter, too: that is the matter of inversion of deposits in lakes, with turnover and mixing. Once it goes down, it doesn't stay down. There would be a different situation at different times of the year.

SETTER: I have a comment to make relative to what happens to fallout material which is brought down with the rain and is, therefore, a container of cistern water supplies. The studies along these lines have been made at the Public Health Service Robert A. Taft Sanitary Engineering Center in Cincinnati some years ago. First of all, the gross beta radioactivity in rain from fresh fallout is largely particulate material which sediments quite rapidly. A proportionate amount of Sr^{90} in mixed fission product is contained in this insoluble material. Older fallout material, probably due to the smaller size of particles and greater surface area, appears to be much more soluble, particularly with respect to the Sr and Cs fractions. Thus, instead of something like 10% being soluble in fresh fission products, up to 50% of old radioactivity falling in new rain will pass through a membrane filter.

In our cistern water studies, the bulk of the fresh fallout will be in the sediments. The sediments, containing organic and microbial activities, result in a low oxidation-reduction potential and an increased dissolved radioactivity in the sediment water, as compared to the overlying clear cistern water. However, this dissolved radioactivity is quite small compared to the total radioactivity in the sludge. It may be that the solubilized fraction in sludge is richer in some specific nuclides of mixed fission products.

Norman E. POLSTER: In operations that involve dairy animals and also vegetable truck farming, has there been any study made of concentration of radioactivity by dairy animals through excreta and manure used to fertilize vegetable gardens? Have any studies been made to show what the relationship is between the truck farm soils and the pasture area?

ALEXANDER: I know of no studies specifically with that in mind. As a matter of fact, there aren't very many of our truck soils that get manure any more.

POLSTER: That is a small farmer problem, then. I was talking about the dairy farmer.

ALEXANDER: The diary farmer doesn't see his manure, it goes back on his own land.

POLSTER: That is what I am talking about, the farmers who put it back on the vegetable farm.

ALEXANDER: Of course, they would be putting some back on. I can give you a few figures about the manure. We conducted an experiment at Cornell University in which we controlled the intake and measured the retention of Sr^{90}. Pigs fed on alfalfa containing Sr^{90} retained 50% of the Ca that went through their gut. They only retained 9% of the Sr^{90} that went through their gut. In other words, there was discrimination by a factor of about 5. So in a way you can say the manure was enriched in Sr^{90} with respect to the hay they consumed. The same thing was done with sheep, except that

these particular sheep utilized a lower percentage of both Ca and Sr^{90}. If you have a discrimination factor of 5 then, relative to Ca, the manure is somewhat enriched in Sr^{90} relative to the hay. But even if you took all of the alfalfa produced on a given piece of land in a year, and then converted that to manure and put that on another piece of land, you still wouldn't be adding a great deal of Sr^{90} with respect to the amount that is already there.

POLSTER: You are talking about equal areas, but you usually don't talk about equal areas.

ALEXANDER: It is a direct proportion. There has been some discussion of that by the British, particularly in one of the Rome conferences (Food and Agriculture Organization of the United Nations). I believe there was some discussion of the use of sludges and various things that had radiostrontium in them, where they were adding quite large quantities to areas close in to waste disposal plants. It is a way of possibly doubling or tripling, the radiostrontium on the ground, I suppose.

DISTRIBUTION AND CYCLING OF FALLOUT NUCLIDES

PART 2. THE TERRESTRIAL ENVIRONMENT
PART 3. THE MARINE ENVIRONMENT
PART 4. THE FRESHWATER ENVIRONMENT

Session Chairman: Alfred W. Klement, Jr.
Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission

SUMMARY OF SESSION, FIRST NATIONAL SYMPOSIUM ON RADIOECOLOGY 1961, CYCLING IN THE TERRESTRIAL ENVIRONMENT*

Stanley I. Auerbach
Ecology Section, Health Physics Division
Oak Ridge National Laboratory**
Oak Ridge, Tennessee

The radioecologist, like other radiobiologists, is concerned about the short and long term impact of radioactivity on the biological or ecological systems which comprise the biosphere. His concern stems not merely from an academic interest in the impact of a powerful evolutionary force such as ionizing radiation, but from his concern about the effect of radioactivity on these ecosystems in terms of their importance for man's well being and economy.

In addition to the more indirect ecological and other biological problems there also is the immediate problem of ecological manipulation of radionuclides which may make radioactive contamination more accessible to the food chains directly serving man. The initial distribution of radionuclides in the biosphere, their subsequent cycling and accumulation, and their ultimate fate, are dependent upon many ecological factors. Our ability to predict the behavior of radionuclides in the terrestrial environment is dependent upon our knowledge of the biological and biogeochemical mechanisms which serve to move and otherwise affect the distribution of chemical elements in the environment.

Cycling of nuclides in complicated ecological systems involves rate processes and transfer mechanisms. Prediction of the behavior of radionuclides in such ecosystems requires that an understanding of these processes be achieved. This situation is analogous to that which existed

* Dr. Auerbach was chairman of Section 1, Session II of the Radioecology Symposium at which the papers reviewed here were presented.

**Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

in physiology before radioisotopes made possible the significant progress in understanding the kinetics of material transfer between tissues and organs in various parts of the body. Formalized approaches involving mathematical models provide a theoretical basis for a systematic attack on ecosystem dynamics and the related problem of radionuclide distribution.

A formalized approach involving the use of analog computers for predicting movement of radioisotopes in ecosystems was described by J. S. Olson (1). His model characterized an ecosystem as a series of compartments which represent the functional organization of the system (Fig. 1). The arrows show the pathways of movement of materials between the different levels of the ecosystem. The wider arrows represent the major pathway of uptake from some exchangeable pool of nutrient material in the soil, to roots, up into plant tops, and then into the photosynthetic parts of vegetation, to be returned to the soil by the pathways indicated. Using this scheme Olson described how analog computers can be used to aid in the interpretation of net changes in radioactivity in parts of an ecosystem.

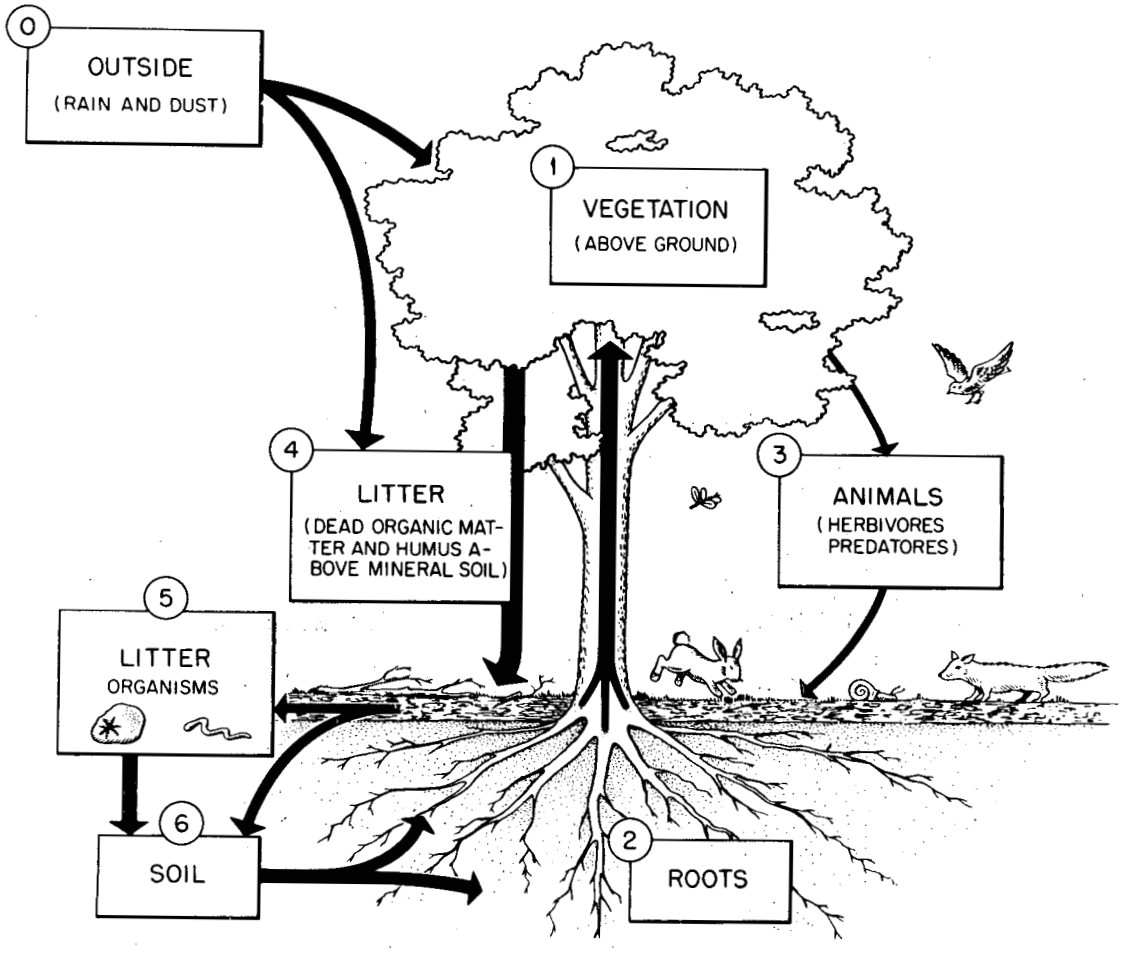
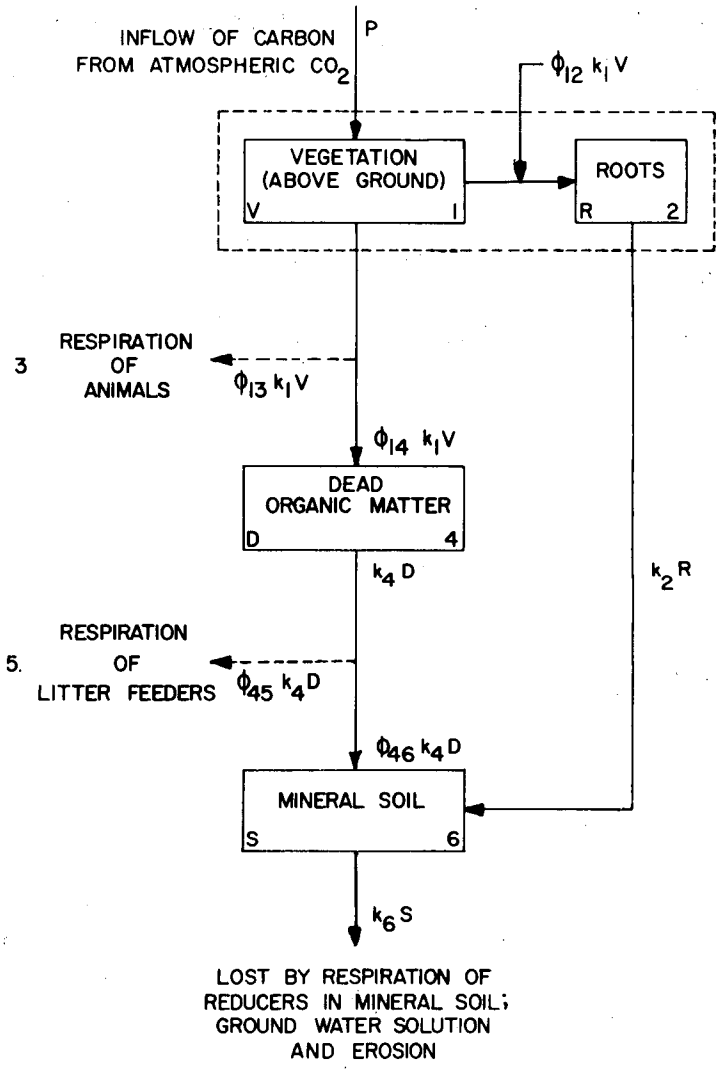


Figure 1. Diagram of a Forest Ecosystem, with Arrows Showing Major Pathways of Transfer of Mineral Nutrients and Radioactive Isotopes Between Compartments. (From Olson (1)).



PARTIAL TRANSFER COEFFICIENTS		DECAY PARAMETERS	
ϕ_{12}	0.20	k_1	0.25
ϕ_{13}	0.00	k_2	0.25
ϕ_{14}	0.80	k_4	0.693
ϕ_{45}	0.50	k_6	0.0156
ϕ_{46}	0.50		

Figure 2. Box Diagram Indicating Pathways of Movement of Carbon in the larger Compartments of a Model Ecosystem. (From Olson (1)).

The basic operation of the analog computer is to keep a running balance of the simultaneous gains and losses for all the major parts or "compartments" of an ecosystem. His paper diagrammatically illustrated how the running balance, or integration, of gains and losses is accomplished. Using carbon as an example, an idealized case was described in which the rate of loss of organic carbon was a constant fraction of the organic carbon accumulated by photosynthetic vegetation; and in which the standing crop of vegetation increased until losses equalled input and a steady state was attained. A block diagram (Fig. 2) showed the pathways of transfer. For carbon, this diagram indicates how one fraction of carbon in the above-ground portion of vegetation is delivered to the plant roots (R); and the other fraction is considered to be consumed by animals, which are not shown as separate boxes for the sake of simplicity of illustration. A third fraction (D) accumulates as dead organic litter. Both roots and litter in turn serve as inputs for the organic carbon or humus in the mineral soil.

Instead of the rather unrealistic assumptions, such as of constant rate of photosynthesis, various elaborations have been introduced to this model one at a time. For example, Fig. 3 illustrates and input which oscillates to simulate the change in production between summer and winter. Furthermore, the mean annual rate of photosynthesis is allowed to vary instead of being kept as a constant over a period of years. Sinusoidal terms are evident in the graph for the vegetation compartment, though somewhat damped in amplitude compared with the oscillations in photosynthetic rate. Each additional stage of transfer, such as that to roots or litter, further attenuates the amplitude of the oscillatory term, so that graphs for these compartments are virtually identical with those that would be expected for a model with an input having no annual oscillation of photosynthesis. Because of the very slow turnover assumed for soil organic carbon, the use of a compressed scale for both axes was necessary.

Further methods for simulating transfers of radioisotopes in more numerous compartments were indicated. A simple example, from Neel and Olson (2), showed how promptly a sudden increase of C^{14} in the atmosphere as a result of thermonuclear bomb tests might modify the C^{14} content of the other parts of the same model ecosystem considered above (Fig. 4). It was assumed that the "normal" amount of C^{14} produced by cosmic rays had previously been incorporated in the ecosystem in direct proportion to the content of total carbon, during the development of vegetation and litter. The soil humus was assumed to have had its equilibrium content of organic carbon and C^{14} already. At the time indicated by the arrow, the analog computer was switched to the "hold" position and the function generator was switched into the circuit. The function generator then fed in a somewhat higher rate of incorporation of C^{14} (Fig. 4, top) which had previously been set up to correspond to the increase projected by Broecker and Olson (3).

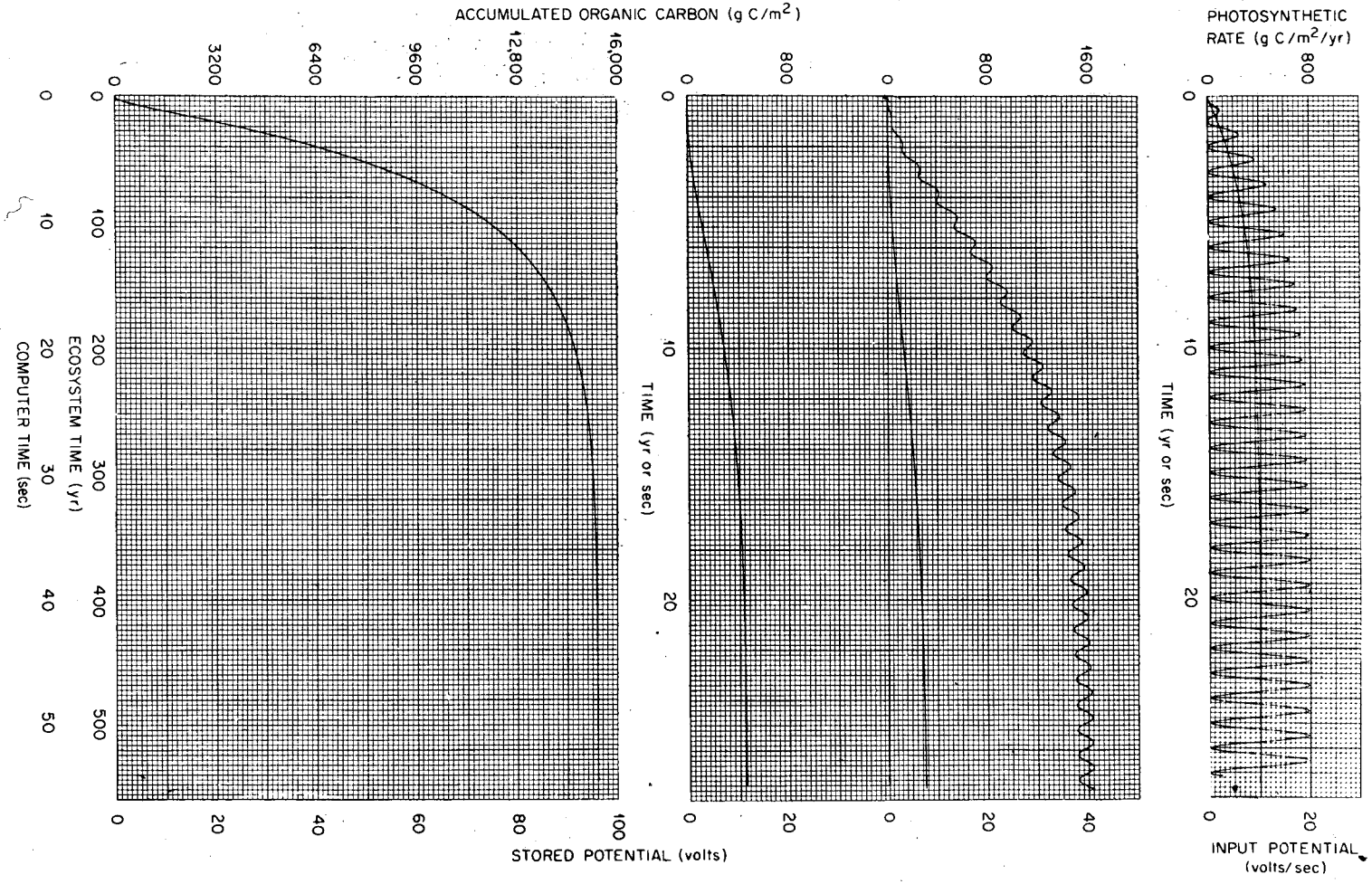


Figure 3. Hypothetical Accumulation of Carbon in a System with an Annual Cycle of High and Low Photosynthetic Rate, Initally Averaging 0 g C/m^2 yr. Gradually Rising to Average of 400 g C/m^2/yr. Sinusoidal oscillations. Sinusoidal oscillations attenuated in Vegetation and almost eliminated in roots and litter under the assumed conditions. Note scale shift for soil organic matter. (From Olson (1)).

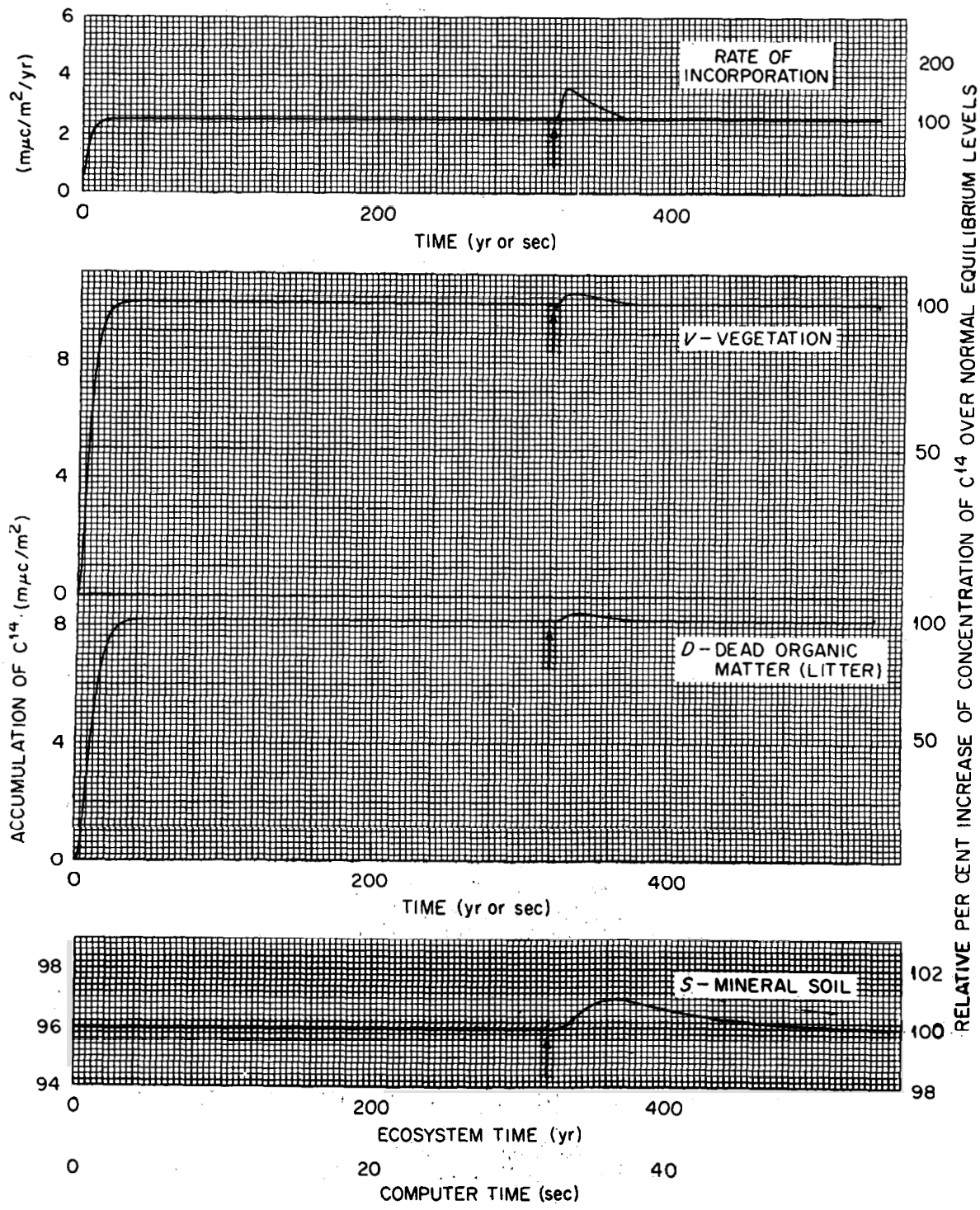


Figure 4. **Response of Model Ecosystem Described in Fig. 2 to Initial** Accumulation of "Normal" C^{14} in Proportion to Total Carbon, and Subsequent Increase in C^{14} Fixation Due to Thermonuclear Bomb Tests. Note lag in response of soil compartment.

Because of the relatively fast time-constants assumed for vegetation and litter, these compartments responded fairly promptly in showing an increase in C^{14}. The slow time constant which was assumed for the turnover of humus incorporated in the mineral soil resulted in a slower response to the "pulse" of C^{14}, and a markedly slower return to the "pre-bomb" levels (assuming that atmospheric nuclear testing and nuclear warfare were not resumed for centuries following the 1958 moratorium on testing). The absolute quantity of C^{14} projected for each square meter of ground surface was somewhat higher for soil humus than for vegetation and litter, but because of the larger storage of carbon, the percentage increase was much less.

Olson pointed out examples of the kinds of ecological conclusions which might follow from more extended analyses of this type, for example: (1) the substantial increase in specific activity of atmospheric C^{14} from weapons tests will have a diluted influence on the specific activity of C^{14} with each transfer through the ecosystem, but the amounts of dilution and promptness of response will vary markedly depending on the parameters of different kinds of ecosystems. (2) Much more widespread atmospheric contamination which might arise in the event of nuclear war would similarly be less spectacular at first than that from fission products, but the long half-life and the capacity for storage of C^{14} in humus as well as in the oceans would mean that any environmental problem involving C^{14} might persist for many generations to follow. (3) The increase in specific activity of carbon already produced from bomb tests provides an unprecedented opportunity for gathering information on the probably holdup of C^{14} in different compartments of the biosphere and for evaluating the consequences of further contamination of this type.

Davis, Hanson, and Watson (4) reported on their studies of the effect of local environmental factors upon accumulation of worldwide fallout in natural populations. They emphasized the importance of local rainfall conditions in conjunction with other ecological factors. Fig. 5 shows the quantities of Cs^{137} in Douglas Fir and Ponderosa Pine foliage plotted against annual precipitation for different rainfall areas in the state of Washington during 1959, the year of maximum Cs^{137} fallout. Although the amount of radiocesium generally rises with increased rainfall there is a decrease in the ratio at areas having greatest rainfall, such as at the Quinalt station in the Olympic rain forest. According to them, this decrease is probably caused in part, by the heavy rains washing radioactive materials from the surface of the vegetation. It would be interesting to confirm this hypothesis by collecting crown entrained with trunk and ground collectors. In this way the total fallout impacted on the crown could be determined as well as the portion transferred to the ground.

Davis, et al. reported another study in which the quantities of gamma emitting nuclides derived from fallout were compared for samples of perennial grass clipped from lowland and upland habitats, both of which

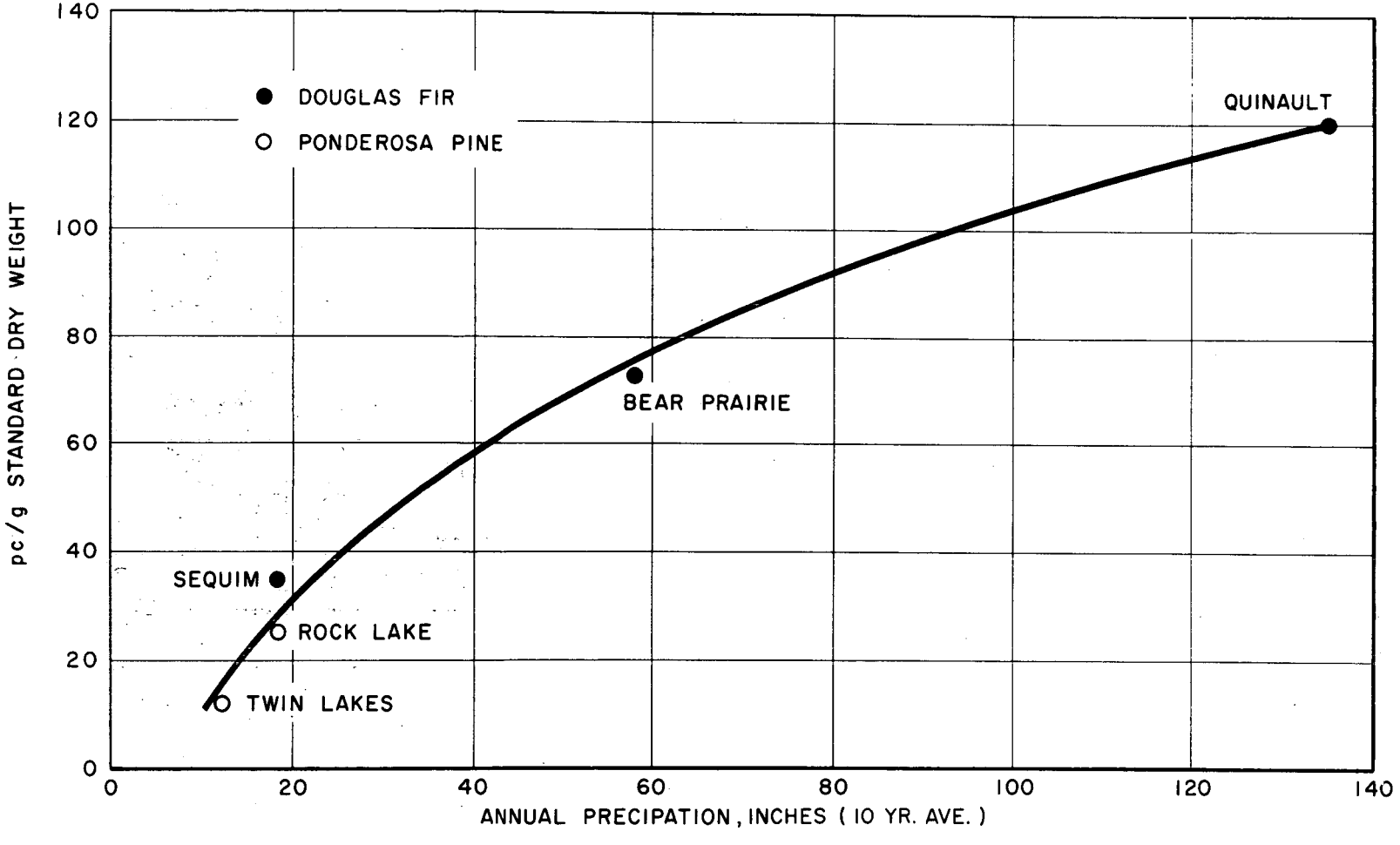


FIGURE 5. Abundance of Cs^{137} in Needles and Twigs of Conifers Relative to Rainfall (From Davis, Hanson, and Watson (4)).

TABLE 1

Comparison of Gamma Emitters in Grass From Lowland and Upland Habitats
Collected at Sequim, Washington, September 18, 1959*

Radionuclide	μc/g Standard Dry Weight				μc/m^2			
	Lowland		Upland		Lowland		Upland	
	mean	std. dev.	mean	std. dev.	mean	std. dev.	mean	std. dev.
Ce^{144}-Pr^{144}	3.4×10^{-5}	$\pm 8.2 \times 10^{-6}$	1.9×10^{-5}	$\pm 4.7 \times 10^{-6}$	2.2×10^{-2}	$\pm 1.1 \times 10^{-2}$	1.4×10^{-2}	$\pm 3.8 \times 10^{-3}$
Cs^{137}	1.6×10^{-6}	$\pm 9.0 \times 10^{-8}$	6.0×10^{-7}	$\pm 1.5 \times 10^{-7}$	1.0×10^{-3}	$\pm 3.1 \times 10^{-4}$	4.3×10^{-4}	$\pm 1.1 \times 10^{-4}$
Ru^{106}-Rh^{106}	2.7×10^{-6}	$\pm 6.6 \times 10^{-7}$	7.3×10^{-7}	$\pm 8.8 \times 10^{-7}$	1.7×10^{-3}	$\pm 7.8 \times 10^{-4}$	5.4×10^{-4}	$\pm 6.4 \times 10^{-4}$
Zr^{95}-Rb^{95}	7.2×10^{-6}	$\pm 1.8 \times 10^{-6}$	4.3×10^{-6}	$\pm 1.2 \times 10^{-6}$	4.7×10^{-3}	$\pm 2.4 \times 10^{-3}$	3.1×10^{-3}	$\pm 1.0 \times 10^{-3}$
Zn^{65}	2.2×10^{-7}	$\pm 3.8 \times 10^{-7}$	not detectable		1.1×16^{-4}	$\pm 1.9 \times 10^{-4}$	not detectable	
Mn^{54}	5.4×10^{-7}	$\pm 8.6 \times 10^{-8}$	1.3×10^{-7}	$\pm 1.3 \times 10^{-7}$	3.4×10^{-4}	$\pm 1.1 \times 10^{-4}$	9.8×10^{-5}	$\pm 1.1 \times 10^{-4}$

* From Davis, Hanson, and Watson (4).

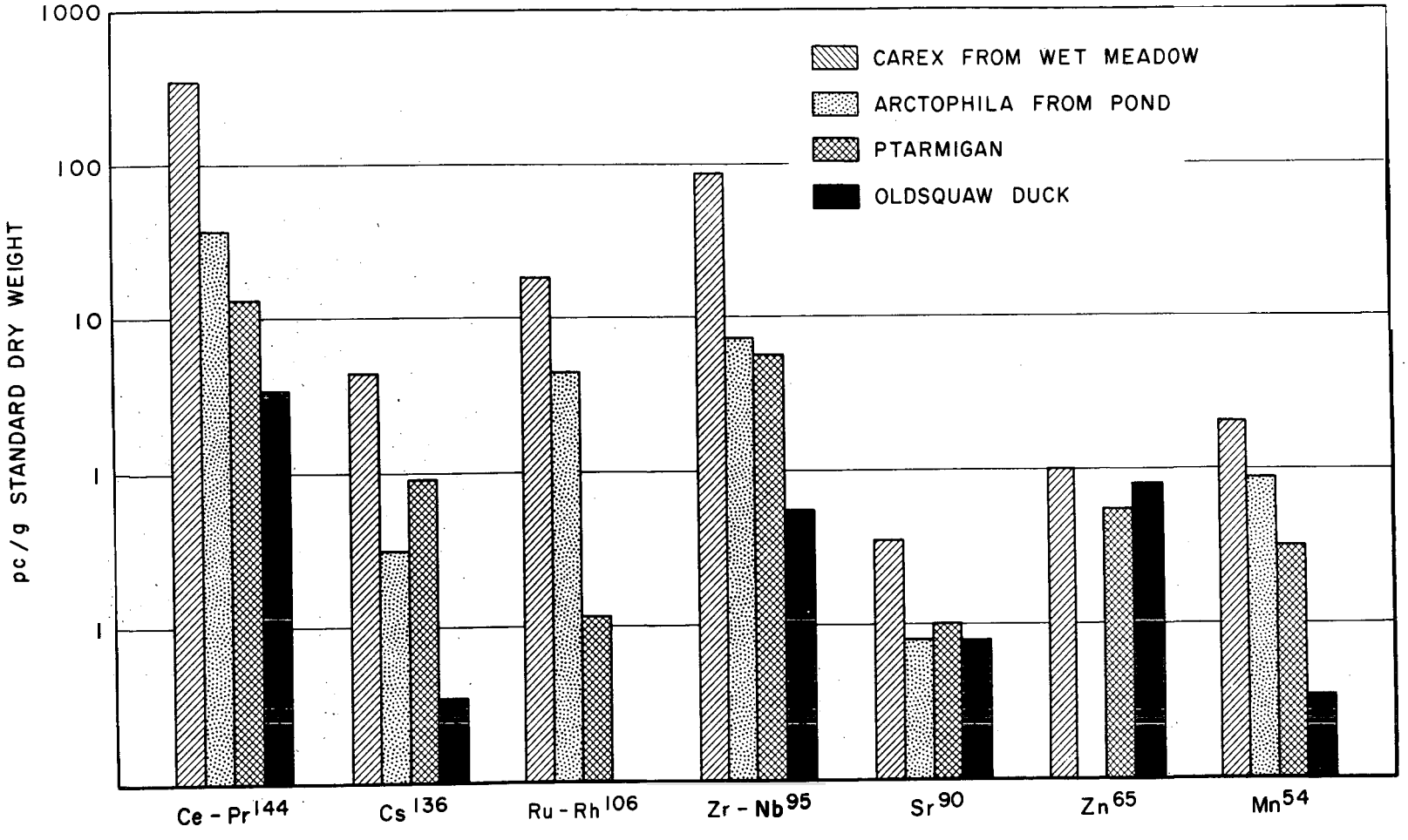


FIGURE 6. COMPARISON OF RADIONUCLIDE CONTENT OF ORGANISMS FROM TERRESTRIAL AND AQUATIC HABITATS (FROM DAVIS, HANSON, AND WATSON (4)).

TABLE 2

Gamma Emitters in Cheatgrass from Burned and Unburned Areas*

Radionuclide	Burned				Unburned			
	μc/g std. dry weight	std. dev.	μc/m^2	std. dev.	μc/g std. dry weight	std. dev.	μc/m^2	std. dev.
Ce^{144}-Pr^{144}	1.4×10^{-5}	$\pm 4.1 \times 10^{-6}$	4.7×10^{-3}	$\pm 1.9 \times 10^{-3}$	5.2×10^{-5}	$\pm 1.5 \times 10^{-5}$	1.0×10^{-2}	$\pm 3.1 \times 10^{-3}$
Ru^{106}-Rh^{106}	7.6×10^{-6}	$\pm 4.7 \times 10^{-6}$	2.6×10^{-3}	$\pm 1.7 \times 10^{-3}$	8.1×10^{-6}	$\pm 3.5 \times 10^{-5}$	1.6×10^{-3}	$\pm 7.7 \times 10^{-4}$
Zr^{95}-Zr^{95}	2.7×10^{-5}	$\pm 3.2 \times 10^{-6}$	9.1×10^{-3}	$\pm 3.6 \times 10^{-3}$	9.9×10^{-5}	$\pm 2.3 \times 10^{-5}$	1.9×10^{-2}	$\pm 5.0 \times 10^{-3}$
Mn^{54}	9.0×10^{-8}	$\pm 1.6 \times 10^{-7}$	4.1×10^{-5}	$\pm 7.2 \times 10^{-5}$	2.6×10^{-6}	$\pm 1.5 \times 10^{-6}$	5.1×10^{-4}	$\pm 2.8 \times 10^{-4}$

* From Davis, Hanson, and Watson (4).

were exposed to like quantities of precipitationaand were geographically separed by a distance of only about 900 m. The lowland grass was salt grass (Distichlis spicata L.). Their data are summarized in Table 1 and show that the radionuclides were most abundant in salt grass whether calculated on a weight basis or total crop harvested per unit area. The reason for the difference is that the impeded drainage afforded a better opportunity for the fallout to be sorbed by the lowland grass. The aerial parts of the plants from both habitats may be considered equally available to direct foliar sorption of radionuclides. However, materials which are deposited in the shallow water or wet soil of the salt grass habitat are more available to the water-exposed parts of plants or shallow roots than those deposited on the soil surface of the upland habitat.

This relationship does not persist, however, as the amount of water of the environment continues to increase, as shown in Fig. 6 which compares the quantities of fallout debris in plants from the wet tundra of arctic Alaska with emergent aquatic plants from a shallow pond (max. depth 8 dm) from the same region. The lower levels of radionuclides in the aquatic plants are attributed to dilution of the fallout by the pond water and removal by plankton organisms. Also compared is the radionuclide content to two species of birds, the ptarmigan which inhabits and feeds in the tundra and the duck which obtains its food from the ponds. The lower levels in the ducks demonstrate the importance of the level of radioactivity in primary producers (plants) upon levels in organisms high in the food chain.

These workers also evaluated the effect of fire upon fallout accumulation in grass. Table 2 summarizes the quantities of radionuclides found in two prairie areas, one of which had been burned in August, 1957, and harvested the following May; and a nearby unburned area. Although the burned area had a greater growth resulting from the release of nutrient elements of the soil with subsequent fertilizing effect, the content of radionuclides was less both on a unit weight and unit area basis than that of the unburned plots. This would indicate that although apparently enough nutrients liberated by the burning were available to thr grass to stimulate vigorous growth, the radionuclides liberated to the soil by the fire were not similarly available. An interesting possibility which this work suggests is the use of controlled burning in heavily contamined areas as an economic means of disposing of badly contaminated crops. In conjunction with soil treatments which also would decrease subsequent availability of crops, such a technique might have merit.

Another interesting aspect of local fallout dynamics was reported by Osburn (5). His work has been concerned with the accumulation of fallout nuclides and their dispersion in high mountain snowbanks in Colorado. These snowbanks are found in the alpine tundra above timberline at approximately 11,000 ft. above sea level (Fig. 7). The study of fallout accumulation in snowbanks may seem somewhat esoteric until it is realized that while the alpine tundra region occupies only about 4%

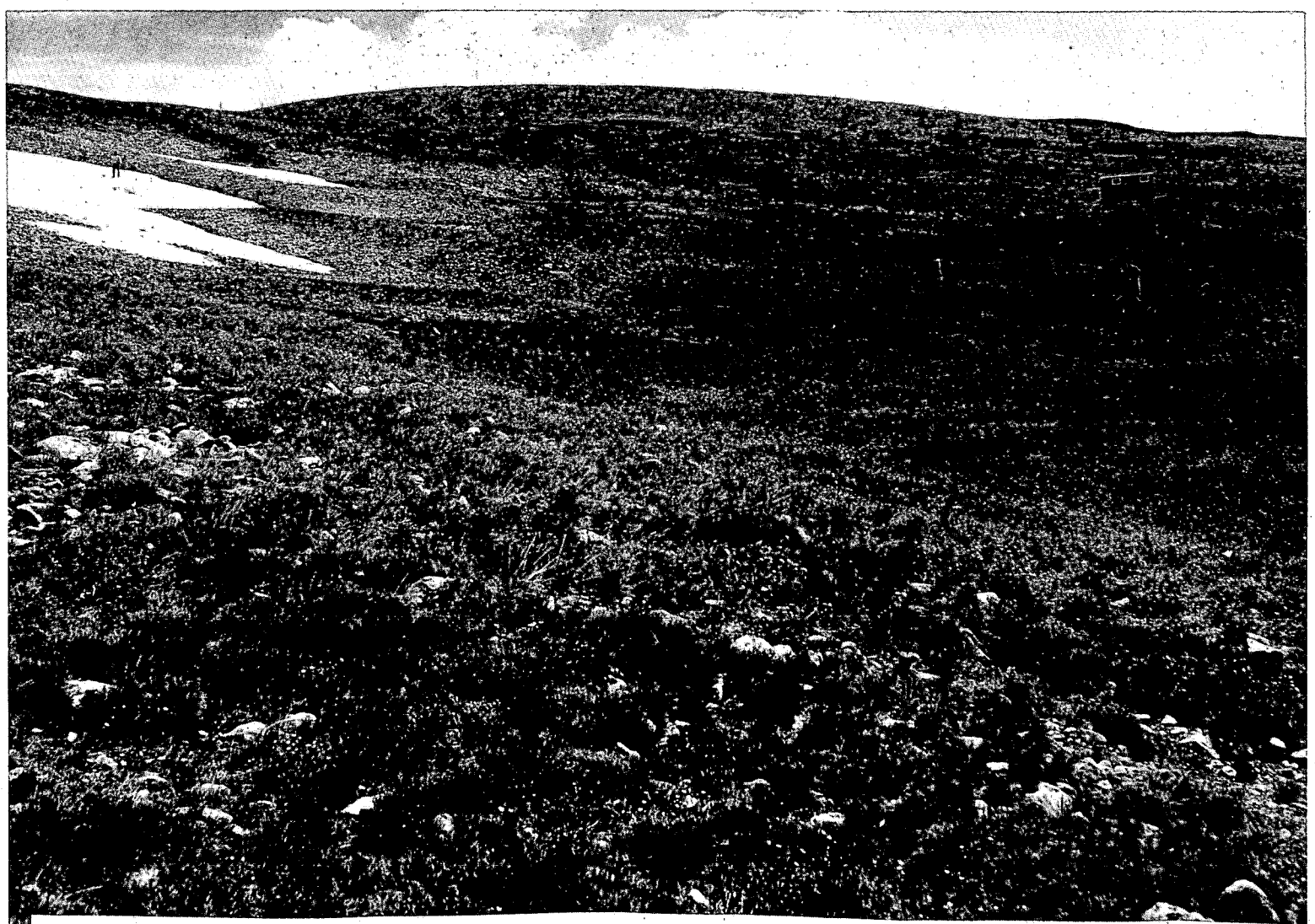


Figure 7. View of snowbank ecosystem on Nicot Ridge (el. 12,000 ft.) which is located about 15 mi. west of Boulder, Colorado, on the east slope of the Colorado front range. (From Osburn (5)).

of the total land area of Colorado, 50% or more of the water utilized for summer drinking and irrigation purposes originates in this region.

At the study site snow covers part of the ground for approximately 10-11 months of the year. The water derived from this accumulation feeds the reservoirs serving the city of Boulder. During the time of snow accumlation, the snowbank accumulates fallout directly from snow, rain, and blow-in of fallout contaminated snow and litter from windward areas. During short intervals, particularly during the spring, easterly upslope winds may occur and frequently they transport dust particles from the grassland plains to situations where they may fall onto or be blown into the snow field. According to Osburn these particles have functioned as collection nuclei for fallout isotopes while they were part of a shifting topsoil and while they were being transported to the snowbank area.

The snowbank with its contained litter and nuclide burden builds up until about mid-May when the snow melting exceeds accumulation. Most of Osburn's numerous samples are still in the process of being analyzed for specific nuclides. But indicative of the results is some data on Sr^{90} determined in samples taken from one of the snowbanks. This particular snowfield had been in existence since 1954. The sample was obtained in October 1960, when the snowfield was reduced to a minimum and had exposed the previous five years of accumulated debris. The debris contained 600 μμc Sr^{90}/g of material. Since each liter of surface snow contains about 1.2 g of debris, each liter of snow-derived water contains approximately 600 μμc of Sr^{90} which is considerably above present drinking water tolerances. Osburn showed that during the meltout period the water percolates down into the snow and finally flows out at the base of the snowbank. During this time the water has relatively little radioactivity because as it percolates through the snow, the radionuclides cling to dust, accumulated litter, and small snow organisms, particularly algae and protozoa. As the snowbank shrinks in mass, the surface becomes more and more radioactive due to the increased concentration of debris.

In those snowbanks where the melt water does contain a high proportion of radioactive debris Osburn showed that this water flow through a sedge mat which grows below the banks. This dense sedge mat and its associated peaty soil functions as a chemical-mechanical sieve which filters out debris and fallout nuclides almost completely. The radioactivity of the snow water may be reduced from several hundred μμc/l to less than one. The practical implications of this type of ecological research is exemplified by these findings. Whereas the alpine snow fields serve as the largest source of late summer water for many towns and cities, nearly all of these snowfields are separated from the collection reservoirs by sedge areas.

The importance of vegetation cover in affecting the impaction of fallout and consequently determining indirectly its availability to plant-feeding animals was also stressed by Rickard (6) in his studies of desert communities at the Nevada Test Site. The influence of life

form and leaf morphology upon the interception and retention of fallout is largely unknown. He suggested that when different plant communities are recognized over the landscape, the plants comprising these communities should be assayed with a representative set of canopy coverage estimates for each community. Such analyses and vegetation maps prepared in advance of fallout events would be of use in estimating the amounts of vegetatively retained fallout by desert shrub communities in the event of widespread fallout occurrences in the shrub dominated regions of the western United States. At Oak Ridge we have long pointed out that forest cover isamong the most effective of ground cover for impaction of fallout because tree canopies consist of many layers of leaves, and trees are superimposed on additional layers of shrubs and herbaceous vegetation.

Two other approaches to the problem of radionuclide cycling in the terrestrial environment were presented at the Symposium. The first was another type of case history approach and the second an experimental approach. For 5 years vegetation studies have been conducted on the radioactive White Oak Lake bed of the Oak Ridge National Laboratory. In this unique area the radionuclides in the soil are present in concentrations which are several orders of magnitude greater than those present elsewhere which are due to weapons fallout. For example Sr^{90} is present in concentrations which range up to 300 mc/acre and Cs^{137} up to 10 c/acre (7). DeSelm and Shanks (8) reported on their 5 years of studies of the vegetational succession on the lake bed which included a study of the accumulation and cycling of organic matter, macronutriet elements, and certain radionuclides. By means of various sampling techniques the annual production of dominant vegetation types was determined during this period. Using these data plus chemical and radioanalyses they have compiled comprehensive information on the cycling of certain radionuclides and macronutrients through some of the dominant vegetation types. These types include woody vegetation represented by the Black Willow (Salix nigra Marsh.), herbaceous vegetation represented by sedge (Carex spp.), rush (Juncus spp.), and Lespedeza. They found that the willow has removed approximately 7% of the total soil Sr^{90}, only 0.028% of the Cs^{137}, and 0.21% of Co^{60}. The herbaceous vegetation removed 1.9% of Sr^{90}, 0.011% of Cs^{137}, and 0.03% of Co^{60}. In terms of annual turnover, assuming constant soil burden and availability, they reported that Lespedeza would return to 76 mc/acre/year, Juncus 25 mc, and Carex 40 mc.

The experimental approach to radionuclide cycling in forest ecosystems was presented in the uaper by Witherspoon (8). He described the first year's results of a study of the cyling of Cs^{134} in white oaks growing on sites of contrasting soil type and moisture. Three white oak trees on each of four soil types were inoculated with 2 mc of Cs^{134}. By a program of sampling and analysis Witherspoon followed the subsequent movement and cycling of this element both on an annual and subannual basis. Movement of radiocesium into the leaves (Fig. 8) continued until about 30 days after inoculation. After this time leaf activity decreased until the end of the growing season when a mean reduction of

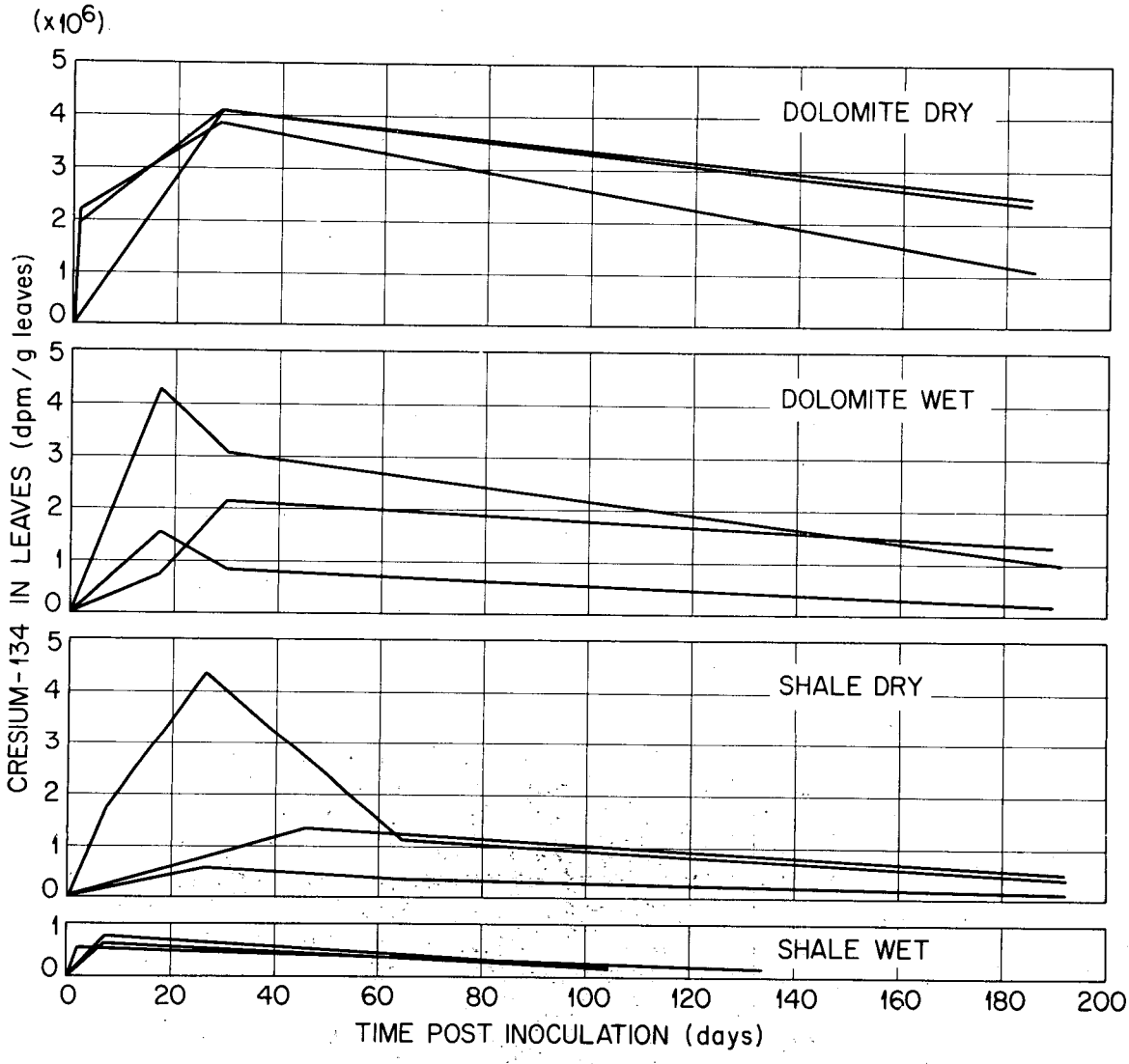


Figure 8. Cs^{134} Gains and Losses in White Oak Foliage During 1960 Growing Season. (From Witherspoon (9)).

63% from maximum was attained. By collecting rain under the crows throughout the growing season Witherspoon showed that approximately 13% of the reduction from leaf maximums was attributable to leaching by rain. The remainder of the difference between leaf maximum and minimum represented radiocesium which had moved back into the woody tissues where it is available for reentry into the leafy portion of the crown the following year.

Radiocesium leached from the crowns moved into the soil profile. Significant differences were found between wet and dry sites in the quantities of Cs^{134} at three soil depths (Table 3). The wet soils had more activity at the 2 to 4 in. level than the dry soils. Radiocesium mobility was greatest in the wet shale soil. The others held over 60% of the activity in the 0 to 1 in. layer.

The rapidity with which a subannual cesium cycle can take place in a forest was also shown by Witherspoon. The soil surrounding small white oak saplings were covered with Cs^{134} tagged leaves. Another series of saplings had Cs^{134} solution applied to the soil. Six days after the application of the tagged leaves and following 1.7 in. of rain, detectable Cs^{134} was measured in the saplings. The rate of uptake of radiocesium by these saplings is compared in Fig. 9. The rate of uptake and accumulation from litter is almost as great as that from the applied solutions. Of the radiocesium that had moved from litter into the top 2 in. of soil, 11% was found to be available for plant use 140 days after application. Witherspoon's work indicates that in natural soil profiles with their intricate structure of roots, worm tubes, pore spaces, radioisotopes are likely to be more mobile than in disturbed profiles.

There are relatively little data on the accumulation of Sr^{90} in bones of native mammals. Skeletal data representing a population or populations are even more meagre. This deficiency is unfortunate because the resumption of weapons testing and the lowering of permissible concentrations as suggested by the Federal Radiation Council and the Public Health Service have given environmental monitoring a new importance. Information on accumulation of radionuclides by wild animals, which occupy various niches in our environments, may be extremely useful in the interpretation of accumulation and distribution of radionuclide contamination in localized situations. The use of organisms as indicators of trends or patterns of environmental contamination has not been fully exploited. In part this deficiency has resulted from a primary emphasis on man and his domestic animals and partly from a lack of understanding and appreciation for the potential value of such research.

A valuable start in this direction was reported by Schultz and Longhurst (10) on accumulation of Sr^{90} in yearling Columbian black-tailed deer. This study was unusual among others of this kind for both domestic and native mammals in that some control over variation due to time, individuals, species, and regions was obtained by using only the jawbones of

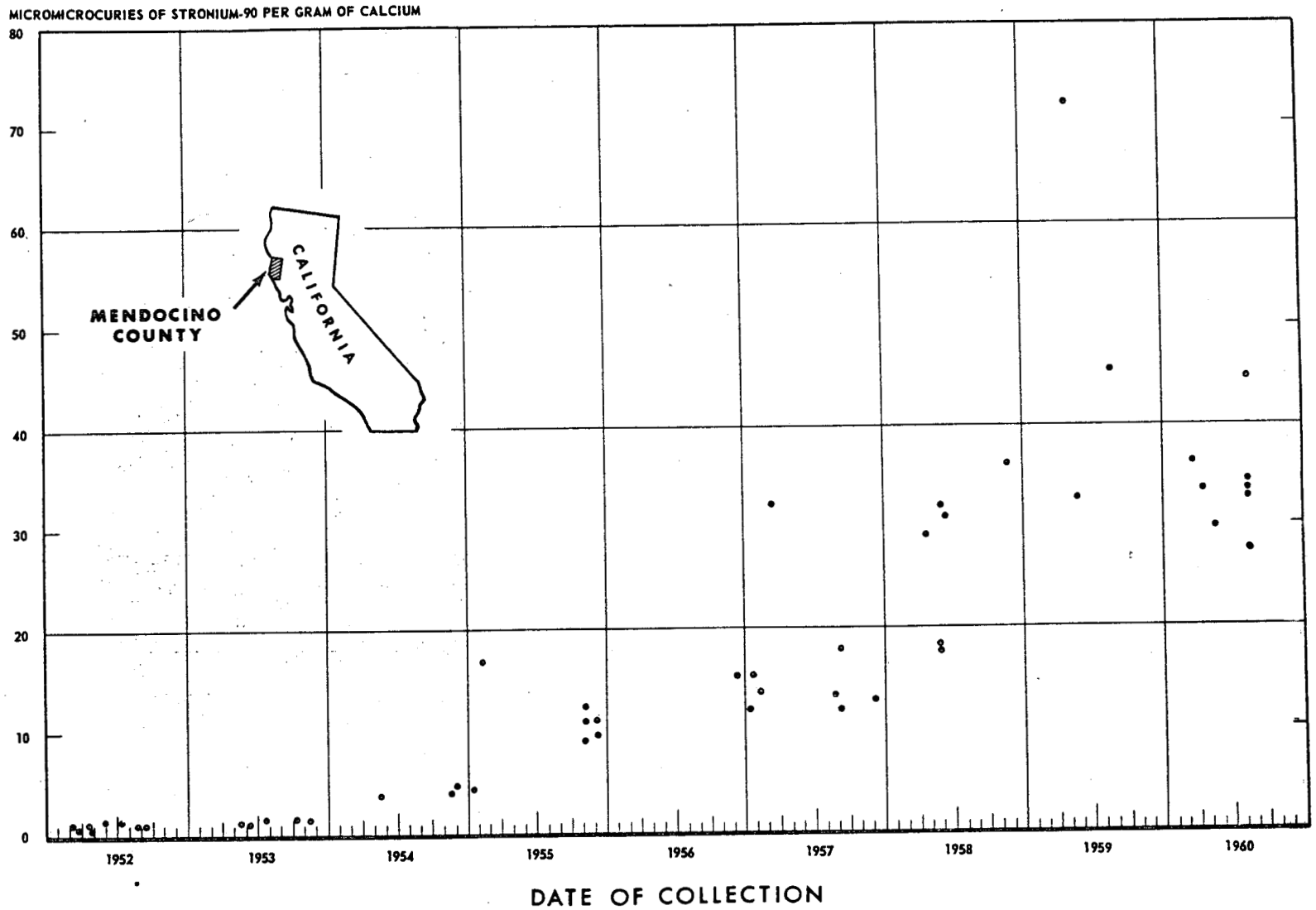


Figure 10. Strontium-90 Content of Mandibles of Columbian Black-Tailed Deer Collected in Mendocino County, California. 1952-1960. (From Schultz and Longhurst (10)).

TABLE 3

Movement of Cesium^{134} Into Soil After Leaching from White Oak Leaves 1960 Crowing Season*

Parent Material	Dolomite		Shale		
Topography	Wet	Dry	Wet	Dry	
Soil Series	Landisburg	Fullerton	Monongahela	Sequoia	Grand Means
% of Activity in 0-1 dm Soil					
0 to 1 in.	65.3 ± 0.43	72.4 ± 7.01	28.5 ± 2.77	68.7 ± 1.51	58.7 ± 10.71
1 to 2 in.	12.8 ± 1.00	20.6 ± 7.33	37.7 ± 2.48	26.1 ± 2.82	24.3 ± 6.07
2 to 4 in.	21.9 ± 0.98	7.0 ± 0.36	33.8 ± 2.53	5.2 ± 1.23	17.0 ± 6.97

* From Witherspoon (9).

yearling individuals from a head maintained in one county in California, representing the same subspecies and collected from 1952 through 1960. Fig. 10 shows the pattern of accumulation of Sr^{90} over this period of time. The data nicely shows the trend of increased Sr^{90} accumulation and suggest a plateau developing following cessation of testing. This species shifts its food habits during the year but unfortunately there are no data on the radiostrontium content of the diet to compare bone content. In view of the statistical control which can be maintained in sampling this mammal further work along these lines seems warranted with renewed weapons testing.

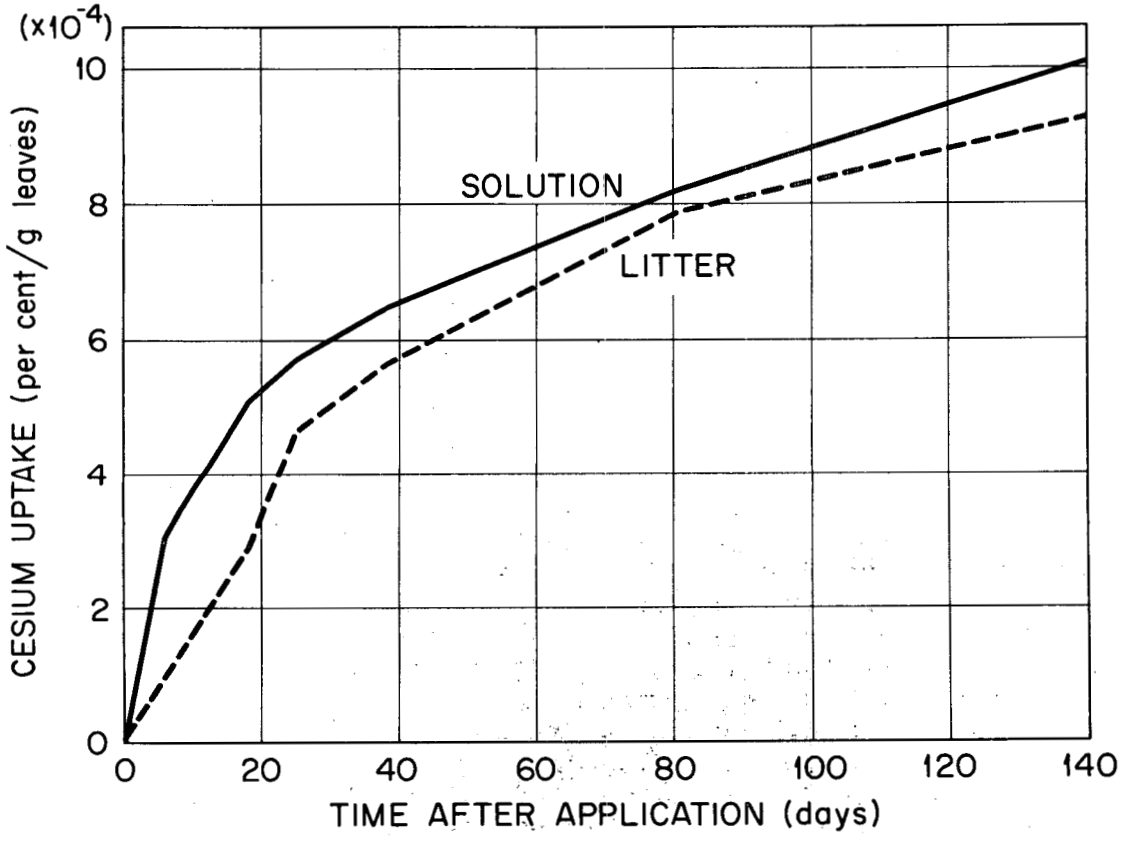


Figure 9. Uptake of Cs^{134} by White Oak Saplings from Tagged Litter and Solution Applied to Soil. (From Witherspoon (9)).

A related study was that of Neel and Larson (11) on the biological availability of Sr^{90} to small native mammals. The source of Sr^{90} in this study was the more localized fallout resulting from devices fired at the Nevada Test Site. Here an attempt was made to use kangaroo rats and jack rabbits as native biological indicators. To test this possibility and in some cases, kangaroo rats were determined in land areas at varying distances from the Nevada Test Site. These areas were in the path of fallout from specific test shots performed during the period of testing. Detailed studies and analysis of the soil showed Sr^{90} to be persistent in the surface soil over a period of one year or more. Despite a rather constant soil level of Sr^{90}, sharp drops in jack rabbit bone concentrations were observed in one study area in 1955 and in another area in 1961. If the bone levels of jack rabbits collected in the latter area in 1958 and 1961 are plotted as a function of body weight, which may be used as an approximate indication of animal age, the values are distributed as shown in Fig. 11. In 1958, one year after the Operation PLUMBBOB "Smoky" shot contamination, 41 of 43 animals had Sr^{90} concentrations in excess of 10 μμc/g Ca regardless of weight (or age). In contrast, in 1961, 39 of 53 animals had bone levels less than 10 μμc Sr^{90}/g Ca. The higher levels were restricted to the heavier, presumably older animals. These data suggest that the higher levels of Sr^{90} are associated with animals which were living early inthe sequence of contamination, that is, the 1957 PLUMBBOB fallout; rather than with rabbits which were born later in an already contaminated environment. There are at least two reasons for this difference in levels. One, plant foilage which serves as food for the rabbits tends to have more contamination immediately after fallout from a shot. Two, with the passage of time biological available Sr^{90} tends to be diluted in the soil mass, reducing both its chemical availability as well as its physical availability. The latter would be important in affecting uptake during dust baths and fur preening.

Reference to dust baths and preening serves as a logical introduction to the paper by French and Larson (12) on the environmental pathways for uptake of I^{131}. These workers pointed out that ingestion is the only method by which significant quantities of radioiodine gain entrance to the animal body under field conditions. This conclusion was derived from several kinds of experiments in which jack rabbits and laboratory rabbits were tested for both inhalation and ingestion of radioiodine. Under field conditions the time it takes for the thyroid to reach maximum activity varies with the species of animal; for the jack rabbit it is the fifth day, the kangaroo rat the tenth day, and for the sheep the twelfth day after ingestion.

By a series of ingenious experiments French and Larson showed that kangaroo rats may obtain radioiodine not only directly from ingestion of contaminated food, but indirectly from their contaminated substrate through food which had been in contact with the substrate, and by

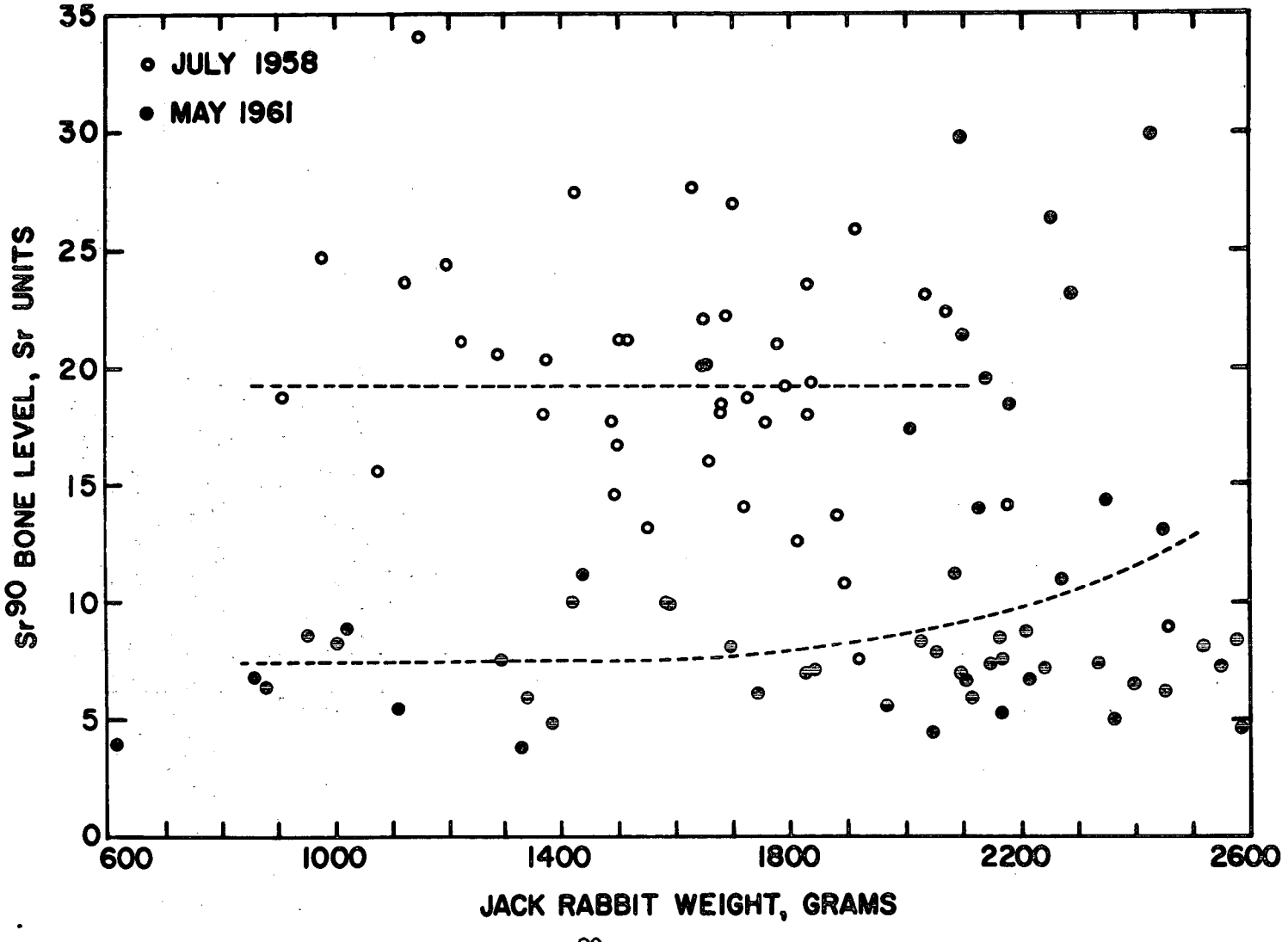


Figure 11. Comparison of bone Sr^{90} levels in jack rabbits collected near PVI Area in 1958 and 1961. (From Neel and Larson (11)).

ingesting contaminated sand during the process of licking and cleaning their fur after a dust bath. In applying their data to a field situation these workers pointed out that fallout is not deposited in the environment on a specific weight basis, rather it deposits on surfaces so that relationships between feeder and substrate should be considered on an area basis. Using this basis they recalculated the respective contribution of each source to the thyroid burden and found that the sand accounted for 1/3 of the body burden of kangaroo rats.

Another aspect of nuclide levels and cycling in relation to animals was provided by two studies dealing with animals on the heavily contaminated White Oak Lake bed. The paper by Crossley (13) on movement and accumulation of radiostrontium and radiocesium in insects in unique in that this radioactive waste disposal area is the only place in which the insect components have been investigated as to their position in the accumulation of radioisotopes in terrestrial ecological food chains. Terrestrial insects are no longer a significant part of a human food chain, but in natural communities they are herbivores of major significance. In aquatic systems, more information concerning the accumulation of radionuclides is available. However, the well known ability of many aquatic insects to concentrations direct from or exchange them with the surrounding medium makes an analysis of food relationships difficult. Consequently, these results of insect studies on White Oak Lake bed may be the only available basis for generalization to other areas which may receive heavy contaminations of Sr^{90} and Cs^{137}.

In an area such as the lake bed the herbivorous and predaceous insects, the plants, and the soil together constitute a system through which the radionuclides are transferred and in which various steady state equilibria may be expected. Fig. 12 shows the concentrations of Sr^{90} and Cs^{137} in organisms at the different ecological levels and in soil. Cs^{137} was more abundant in soil than was Sr^{90} but was less available to plants, so that Sr^{90} concentrations in plants were higher than Cs^{137} values. Sr^{90} was concentrated above soil levels in plant leaves. The herbivorous insects contained about equal quantitites of the two radionuclides, since radiostrontium is eliminated more rapidly than radiocesium and equilibrium values thus were much lower than were concentrations in the host plants. The predaceous insects had approximately the same concentrations as did herbivorous insects, but were represented by only a few samples.

When the actual biomasses of the ecological levels involved are included (Fig. 13) reduction in transfer of radionuclides from one level to another becomes the rule. The mass of soil overwhelms the masses of the organisms, so that the soil contained more than 99% of the radionuclides in the system, Although this food chain system shows reductions at each major step, this does not necessarily apply to other chains. Birds living on White Oak Lake bed had higher concentrations of Sr^{90} than was found in the insects upon which they feed, due to accumulation in bone. The concentration of radionuclides (specific wt. basis) in the food

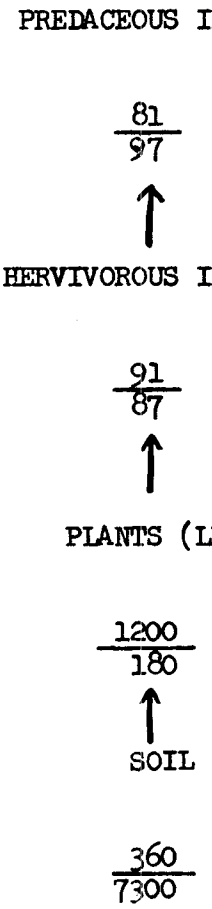


Figure 12. Concentrations of radionuclides in soil and organisms of the White Oak Lake bed (upper value Sr^{90}, lower value Cs^{137}, in μμc/g dry wt.) (From Crossley (13)).

chain will depend upon the average elimination constants or concentration factors in each step involved. When the amounts of radionuclides in the ecological levels (area basis) are considered, vast deductions occurred during the transfer of radionuclides from one ecological level to another.

Evidently the insects have very little effect on the distribution of radionuclides on the lake bed or in removal of nuclides from the system. In the extreme case, Crossley calculated that if all the insects on the lake bed at any one time were to leave, the loss or radionuclides would be about 3 μc each of Sr^{90} and Cs^{137}.

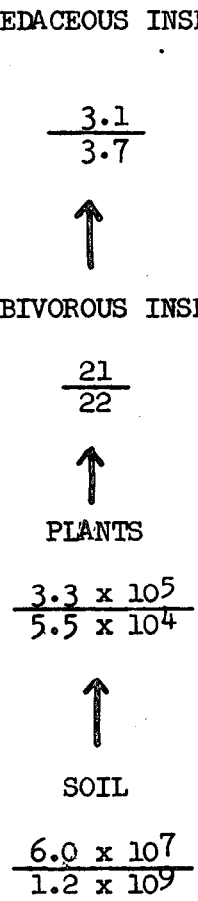


Figure 13. Distribution of radionuclides in soils and trophic levels of the White Oak Lake bed (upper value Sr^{90}, lower value Cs^{137}, in $\mu\mu c/m^2$). (From Crossley (13)).

Kaye and Dunaway (14) have been conducting population studies of cotton rats which are one of the native field rats on White Oak Lake bed. They have also been determining the body burdens of radionuclides in these rats which are resident and feed on the contaminated vegetation of the bed. They demonstrated how the body burdens of two rats could be used as examples for calculating whole body dose rates in native animals from radionuclides deposited in tissues and in the gastrointestinal tract (Table 4). Using the data on these known age rats who were marked as young litter mates and released on the bed and applying modifications of standard equations they obtained the dose rates shown in Table 4. Of the dose delivered by the internally deposited nuclides, Sr^{90} accounts for over 80%. The greatest portion of the integrated total dose is derived from external exposure to the radiation field over the lake bed.

TABLE 4

Burdens of radionuclides in two cotton rats from White Oak Lake bed*.

Animal Number	Age (days)	Animal Fraction	Burden of Radionuclides ($\mu\mu c$) Sr^{90}	Ru^{106}	Cs^{137}	Co^{60}
156	170	Whole Body (Exclusive of GI Tract Cont.)	12,800	1,300	1,090	616
		Contents of GI Tract	259	2,250	2,010	1,500
158	170	Whole Body (Exclusive of GI Tract Cont.)	9,420	1,210	998	598
		Contents of GI Tract	288	587	1,790	1,140

*From Kaye and Dunaway (14).

TABLE 5

Estimated dose rates for two cotton rats from White Oak Lake bed*.

Whole-Body Dose Rates (rem/wk.)

	Source of Estimate				
	A	B	C	D	E
Animal Number	Residual Carcass	GI Tract with Contents	A + B	External Sources	C + D
156	0.30	0.08	0.37	2.5	2.9
158	0.27	0.04	0.31	2.5	2.8

*From Kaye and Dunaway (14).

Kaye and Dunaway also presented parallel data on radionuclide accumulation in muskrats which occupy a radioactive waste settling basin at Oak Ridge National Laboratory. Using these data (Table 6) they illustrated how dose equations may be applied to these accumulations. The dose rates obtained are summarized in Table 7. The radionuclides present in the gastrointestinal tract may add significantly to the whole body dose. For specimem 202 (Table 7) the relative increase in dose rate is slight when the gastrointestinal tract contents are included, but for the other three specimens the increase is larger especially for specimen 204 which is increased seven-fold from 15.1 rem/wk. to 112 rem/wk. The true dose rates probably lie between the two values calculated for each muskrat and would be high enough to suggest that radiation induced pathologies or premature aging might result from such dose rates if the animals were exposed continuously at this level. Gross pathological examinations of all four animals and diagnostic x-ray films of Nos. 279 and 280 did not reveal any unequivocal lesions. However, it should be pointed out that the time of exposure to radiation was not known for these animals and may have been insufficient to cause effects.

E. P. Odum (15) presented a paper that was a parallel to that of Witherspoon (9) in that it dealt with the use of radioactive tracers for deriving experimentally certain ecological relationships. In this work P^{32} was applied to three species of plants which comprise an important vegetation component of an old-field ecosystem. Subsequent to application the quantities and distribution of radiophosphorus were determined in nearby animal populations. When the concentration of the tracer per unit of biomass was plotted against time, a clear graphic separation of certain feeding levels and habitat groups was evident for all three plant species.

The trophic position of ecological feeding level of two of the most common animal species, whose exact food source in nature was previously unknown was rather clearly established by comparison of their uptake curves with those of species whose food was known. Odum's experiments demonstrated that the radioactive tagging of single species of producer organisms can aid in the isolation or "untangling" of food chains, as well as aid in the determination of the energy sources being utilized by specific heterotrophic populations in nature.

TABLE 6

Body burdens of radionuclides for muskrats from ORNL settling basin*

Animal Number		Burden for Radionuclides (μc)					
		Cs^{137}	Co^{60}	Ru^{106}	Zn^{65}	Ce^{144}	Sr^{90}
202		0.639	0.859	0.394	*	*	1.370
204	GI Tract	0.976	67.600	*	*	*	12.400
279	W/Cont.	0.122	0.089	0.014	0.560	0.077	0.214
280		0.478	0.207	0.045	trace	0.285	0.174
202		3.710	0.098	*	0.053	*	13.800
204	Residual	1.140	0.122	*	0.193	*	8.370
279	Carcass	0.312	0.071	0.152	0.274	0.116	8.790
280		2.040	0.094	0.450	0.029	0.054	6.540

* From Kaye and Dunaway (14).

TABLE 7

Estimated dose rate for four muskrats from a settling basin for radioactive liquid wastes*

Animal Number	Whole-Body Dose Rates (rem/wk.) Source of Estimate		
	A	B	C
	Residual Carcass	GI Tract with Contents	A + B
202	23.7	4.1	27.8
204	15.1	96.9	112
279	13.2	8.0	21.2
280	8.3	9.5	17.8

* From Kaye and Dunaway (14).

Summary Comments

Information on the ecological factors which affect the initial distribution of radioactive contamination in the biosphere, its subsequent circulation and accumulation, and its ultimate fate, is essential for judging whether radioactivity from fallout would affect natural ecosystems or would be transferred through food chains leading ultimately to man. Differences in vegetation cover and accumulation will affect the quantity and availability of fallout nuclides to plant feeding animals. There are relatively little systematic data on the accumulation of radionuclides by native animals. Information on accumulation of radionuclides by wild animals, which occupy various niches in our environments, may be extremely useful in the interpretation of accumulation and distribution of radionuclide contamination in localized situations.

The understanding of the biogeochemistry of natural major and minor elements in ecosystems is closely related to the interpretation of the movement of radioactive contamination through these systems. The possibility of such contamination arising from radioactive waste disposal, weapons fallout, or perhaps from nuclear war, leads to the need for mathematical models of conditions which could predict the extent and time scale of such contamination. The formulation and testing of such models must draw upon tracer experiments and other basic research on the processes taking place in ecosystems.

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15. Odum, E. P. and E. J. Kuenzler. 1961. Experimental Isolation of food chains in an old-field ecosystem with the use of P^{32}. University of Georgia. Symposium Paper No. 16.

*Papers presented in Session II: Section 1, Cycling and Levels of Nuclides in the Terrestrial Environment. First National Symposium on Radioecology, Colorado State University, Ft. Collins, Colorado, September 10-15, 1961. (Proceeding to be published, U. S. Government Printing Office. V. Schultz and A. W. Klement, Jr., eds., 1962).

SUMMARY OF REPORTS ON FALLOUT NUCLIDES IN THE MARINE ENVIRONMENT PRESENTED AT THE 1961 RADIOECOLOGY SYMPOSIUM

Allyn Seymour*
Laboratory of Radiation Biology
University of Washington

The reports summarized here were presented in Part A:, "The Marine Environment," of Session II: Section 2, "Cycling and Levels of Nuclides in the Marine and Freshwater Environments," of the First Symposium on Radioecology. The symposium was convened September 10-15, 1961, at Colorado State University, Fort Collins, Colorado, under the auspices of the American Institute of Biological Sciences and was sponsored by the Environmental Sciences Branch, Division of Biology and Medicine, U. S. Atomic Energy Commission, in cooperation with the Institution of Environmental Biology at Colorado State University. Dr. T. R. Rice acted as chairman of Part A, Session II, Section 2, in the absence of Dr. W. A. Chipman. The papers presented are listed at the end of this paper.

In preparing the summary, note has been taken of the comment regarding the purpose of this conference, namely, "... that the conference be of a technical nature with emphasis on research results," and also of the comment in the guide for preparation of the papers that the review papers, "... should be related to fallout generally..." For this reason, certain papers will receive more attention than others in the summary.

The twelve papers that are reviewed here report the results of both field observations and laboratory experiments that are related to the biological, physical and chemical aspects of fallout **nuclides** in the sea.

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* Presented by Dr. Edward **E**. Held, University of Washington, for Dr. Seymour.

In the latter category is the paper by Bowen and Sugihara (1), "Cycling and Levels of Strontium-90, Cerium-144, and Promethium-147 in the Atlantic Ocean." The authors discuss the levels of these three fallout **nuclides** in various parts of the Atlantic from 1956 to 1960 as well as their ideas about the processes that move these nuclides through the marine hydrosphere.

Bowen and Sugihara report that the distribution of Sr^{90} in the surface waters is more patchy for waters of the continental shelf area than for large areas of the open ocean. In 1956, Sr^{90} values for samples from the continental shelf area ranged from 6 to 30 dpm/100 l. The size of the patches was estimated to extend from 1 to 5 kilometers horizontally and 30 to 50 meters vertically. However, the fact that since 1956 the range of values has decreased each year in spite of an increase in delivery of Sr^{90} to the ocean is evidence of both horizontal and vertical mixing.

The vertical distribution of Sr^{90} in waters beyond the continental shelf was also determined and in 1958 Sr^{90} was found as deep as 2,000 meters. From these data the amount of fallout per unit surface area of the ocean was determined by integration and was found to be significantly greater than terrestrial fallout at comparable latitudes. Two suggestions are offered to explain the difference: (1) either the transfer rate of fallout from the stratosphere to the troposphere is greater over the ocean than over land due to the mechanics of oceanic high pressure cells, or (2) the scavenging in the troposphere of Sr^{90} by precipitation is more efficient over land. The authors have a slight preference for (2) based upon their belief that little Cs^{137} or Sr^{90} is removed from the water and the constancy of the observed ratio of Cs^{137} to Sr^{90} in sea water. Over large areas of the ocean the ratio remains close to 1.2 which is in contrast to a fission yield ratio of 1.5 to 2.0 and a rather consistent ratio of 2.5 to 2.8 both in rain (over land) and in high altitude air. These differences in ratios indicate that the methods of removing fallout from the air are different over land than over sea.

The two lanthanides, Ce^{144} and Pm^{147}, are distributed differently in the ocean both in respect to each other and to Sr^{90}. Maximum values for Ce^{144} and Pm^{147} occurred at different depths and the ratios of one to the other changed constantly. Unlike Sr^{90} these two **nuclides** are removed from sea water by physical and/or biological processes. For these reasons the integration of either Ce^{144} or Pm^{147} values obtained at various depths at one station in order to estimate the fallout per unit area at that station is meaningless.

For reasons that follow the authors are of the opinion that Sr^{90} moves as a solute and its movement indicates the movement of water. Sr^{90} exists in fallout in soluble form and upon reaching the sea surface it begins to mix with large amounts of both stable strontium (9 mg/l) and stable calcium (400 mg/l). Once mixing has taken place a net loss in the water of Sr^{90} relative to stable strontium can take place only by isotopic exchange between the water and a solid and under certain conditions. The conditions are that in a solid there must be a high relative concentration of another

nuclide of the element or a chemically similar element and in a mobile form so that the chances that the Sr^{90} ion, once it has moved from the water to the solid, will return to the water are less than for another Sr^{90} ion in the water to move to the solid; also, to be effective, there must be a relatively rapid cyclic exchange of ions between the water and the solid. If the ion-exchange process is at work in the ocean then the site of the calcitic oolites of the Great Bahamas Bank would be one place where a reduction of Sr^{90} in water would be expected, but there is no such evidence. Removal of Sr^{90} from sea water by biological organisms cannot be great either, because strontium is not strongly concentrated by any organisms but the Acantharia and cannot be thought to be held in isotopic equilibrium with the environment by any organisms, including the Acantharia. The final bit of evidence that leads to the conclusion that Sr^{90} must be involved in the full circulatory cycle of the ocean is that the ratio of Cs^{137} to Sr^{90} is constant. As the two **nuclides** are chemically different, the removal of one could not be related to removal of the other; therefore, the fact that their ratio remains constant suggests that both elements move by circulatory processes only.

If Sr^{90} moves as the water moves eventually the Sr^{90} in deep water will return to the surface. The authors believe the return has begun and that this is the process, although the data are still scant, that explains the sharp and significant jump in surface Sr^{90} values from the open sea in late 1959 and 1960. (For the Sargasso Sea and southwest North Atlantic the change was from about 11 to 24 dpm/100 l; for the subequatorial Atlantic, 6 to 26; and Caribbean, 7 to 14).

"It may well prove to be the first indication that maximum dilution by deeper water moving to the surface is no longer effective, because the levels from which return is most significant have reached surface concentration of strontium-90."

Unlike Sr^{90}, the movement of Ce^{144} and Pm^{147} in the ocean is associated with sinking particles, and of the two elements it has been shown that promethium is removed preferentially and sinks faster. Furthermore, the association of Ce^{144} and Pm^{147} with either organic or inorganic materials in the water is strictly a surface adsorption phenomenon, and if both are present they will compete for cerium or promethium. In conclusion, the authors state that the term "cycle" is not appropriate to describe the distribution of Ce^{144} and Pm^{147} in the ocean, because these two **nuclides** become associated with small, slowly sedimenting particles and pass steadily to the bottom sediments from which they do not return, with the possible exception of back-diffusing phosphate complexes.

Additional information about the role of sediments in removing radio-**nuclides** from ocean waters is proved by Duke et al. (2) in their paper, "Availability of Sediment-Sorbed Materials to Marine Biota." They recognize the commonly accepted viewpoint that the sorptive properties of clay minerals, diatom frustules and other common suspensoids, potentially, provide a means by which radioisotopic ions can be transferred from ocean

water to the bottom sediments and thereby decrease the biological availability of certain radioisotopes; however, they also recognize the possibility that this scavenging process may increase the biological availability of certain radioisotopes. The argument is that sorption is a method of concentration and of transforming dissolved material into solid aggregates; thus when radioisotopes are attached to a solid they may become available to bacteria or to filter-feeding organisms, whereas they may not be available if they remain in the ionic form.

These were the thoughts that motivated the laboratory experiment reported by Duke et al. Specifically, their objectives were to determine if clay associated materials were available to the biota and if clay sediments could be expected to provide a retentive reservoir for radioisotopes scavenged from water.

There were three groups of experiments: in one, the ability of marine bacteria to remove C^{14} from montmorillonite, a clay that had been previously labeled with a C^{14} organic material, was measured. After seven days the amount of C^{14} remaining on the clay in the lots with the bacteria was one half the value for lots without bacteria. This observation was interpreted as meaning either that the bacteria sorbed material directly from the clay particles or, that by using dissolved material from the water they changed the ionic equilibrium between the water and the clay in such a way that it favored the transfer of C^{14} from clay to water. In either case the bacteria were effective in removing C^{14} from the sediments and the authors conclude that one may expect a similar effect with other radionuclides bound in organic materials.

In the second group of experiments fish, shrimp and oysters were exposed to a slurry of C^{14}-labeled algal material sorbed on montmorillonite. The slurry was merely added to the aquaria in which the shrimp and the oysters were living but was force-fed to the fish. There was a significant uptake of C^{14} only by the shrimp.

In the third group of experiments the effect of montmorillonite upon the biological uptake of radioisotopes in solution was measured. The test animals were fish, shrimp and oysters and the **nuclides** were Cs^{137}, Ca^{45} and C^{14}; Cs^{137} and Ca^{45} were in the chloride form, and C^{14} as labeled algal material. In these experiments the accumulation of radioisotopes by the test animals was unaffected by the presence of montmorillonite.

As a result of these experiments the importance of clay sediments in scavenging radioisotopes from ocean waters may be somewhat less than commonly believed. First, the sorption of radioisotopes on clay particles may make some **nuclides** more available to certain organisms. Secondly, the supposed scavenging properties of suspended particles was not demonstrated despite the use of a highly sorptive precleaned clay in proportional quantities far exceeding those usually found in nature.

The horizontal movement of radioisotopes in the ocean is dependent principally upon ocean currents, especially for radioisotopes that are present

as solutes or sorped by plankton. An estimate of the westward movement of radioactive water from the Bikini - Eniwetok area was reported in the paper by Seymour (3), "Radioactivity of Marine Organisms from Guam, Palau and the Gulf of Siam, 1958-1959." The rate of advance (or transport) of fallout by the North Equatorial Current from the Bikini - Eniwetok area to Guam and Palau (which are 1,200 and 1,950 miles, respectively, from Bikini-Eniwetok) was calculated to be eight miles per day. Rates of transport of radioactive water from the Bikini-Eniwetok area based upon other data ranged from seven to ten miles per day. The criterion for determining the arrival of radioactive water at Guam and Palau was a significant increase in the gross beta counts of certain marine organisms. The samples with the highest gross beta counts were Guam samples of clam kidney and spider snail liver, for which the values were 204 and 356 μμc/g (wet), respectively. In addition to naturally occurring K^{40} the gamma-emitting **nuclides** present in the Guam samples, in order of abundance, were Co^{57}, Co^{60}, Mn^{54}, Ce^{144}, Zn^{65}, Zr^{95} and Cs^{137}. The presence of Cs^{137} in a marine organism in detectable quantities was unexpected; however, on another occasion Cs^{137} also was detected in the muscle of spiny lobster.

The papers remaining to be reviewed are reports about environmental factors affecting the uptake and biological cycling of radioisotopes. The paper by Townsley et al. (4), "The Effect of Environmental Ions on the Concentration of Radiocalcium and Radiostrontium by Euryhaline Teleosts," is in the former category. A review of the literature on the absorption, incorporation and excretion of ions by marine organisms leaves the authors with the impression that the results of research at the present time are conflicting, if not contradictory.

The first part of the experiment was designed to determine whether absorption of radiostrontium is a function of only the stable strontium, or the calcium plus strontium, or the total ion content of the water. The test animal used in all experiments was Tilapia mossambica, a fish that lives in either fresh, brackish or sea water. After 16 days the accumulation of Sr^{89} was 24 times greater for fish living in tap water than for fish living in sea water or in tap water to which calcium had been added to equal the calcium in sea water (400 mg/l). There was no Sr^{++} and only 8 mg/l of Ca^{++} in the tap water. The uptake of Sr^{89} by fish living in sea water and in tap water with added Ca^{++} was not significantly different. For all three lots uptake continued to increase during the length of the experiment. From these observations it was concluded that the addition of Ca^{++} reduced the uptake of Sr^{89} but not in proportion to the amount added. A fifty-fold increase in Ca^{++} in water resulted in a twenty-four-fold decrease in the amount of Sr^{89} accumulated by Tilapia.

The objective of the second part of the experiment was to determine the discrimination function for Sr^{90} relative to Ca^{45} by comparing the Sr^{90}/Ca^{45} ratios in water and in fish. ($DF = \frac{Sr^{90}/Ca^{45}, \text{ fish}}{Sr^{90}/Ca^{45}, \text{ water}}$). The

experiment was designed to rear Tilapia in fresh water, natural sea water and artificial sea water in which the Sr^{++} to Ca^{++} ratio varied from lot to lot but in which the Sr^{90}/Ca^{45} ratio remained constant. After three weeks the Sr^{90}/Ca^{45} ratio was less in fish than in water for all lots. This means that relatively less Sr^{90} than Ca^{45} was accumulated from the water or, in other words, there was a discrimination against strontium. By lots the DF values ranged from 0.88 to 0.48.

For the three lots, i.e., fresh-water, natural sea water and artificial sea water in which the Ca^{++} and Sr^{++} concentration was the same-- 400 mg/l, and 8mg/l,--the DF values were 0.88, 0.60 and 0.48, respectively. In other words, for the entire experiment the discrimination against strontium was least in fresh water to which had been added 400 mg of Sr^{++} and 8 mg of Ca^{++} per liter and greatest in artificial sea water with the same Sr^{++} and Ca^{++} concentration. Therefore, it would appear that the author's conclusions should be accepted with reservation; they conclude that the accumulation and retention of radiocalcium and radiostrontium are controlled by the total calcium present, as well as other salts in the environment, and that increased concentrations of Sr^{++} in the environment have little effect upon the total radioactive calcium and strontium absorbed, as well as their ratio to each other, except in the absence of calcium and possibly other divalent ions. The authors also conclude that for decontamination of aquatic environments the addition of calcium salts and other sea water ions would be more effective against absorption of radiocalcium and radiostrontium than the addition of Sr^{++}.

The final experiment was designed to locate within the fish the site at which discrimination against strontium takes place. To do this the fish were placed one at a time in a non-mobile position with a dam separating the anterior from the posterior portion of the fish. Ligation of the esophagus to prevent swallowing also was used in part of the experiment. Although there was no direct evidence as to the particular region where discrimination against Sr^{85} takes place, it was found that the amount of Sr^{85} moving through the fish is nearly equivalent in both the anterior and posterior portions of the fish and that ligation of the esophagus does not produce any marked effect upon uptake. Although Tilapia in fresh water pass about ten times as much water through their epithelium as they do in salt water there is no significant effect upon uptake. The authors conclude that this is of particular importance to the study of radioactive contamination, because it means that ions which follow water movement will most likely become sources of important contamination in the tissues.

Lowman (5) in his paper, "Radionuclides in Plankton and Tuna from the Central Pacific" also mentions that one of the three factors that control the uptake of a specific radioisotope from sea water is the concentration of ions of the same element or chemically similar elements in the water. The other two factors are 1), the tendency of divalent cations to complex strongly with biological substrates; and 2), the biological requirements for certain elements in metabolic processes.

Relative to the accumulation of radioisotopes in plankton, Lowman points out that even though plankton may concentrate radionuclides from the water by a thousandfold the total amount of radionuclides remaining in the water is still considerably greater than in the plankton because the ratio of water to plankton in the area where his samples were collected is about 100,000,000: 1. The kinds of radioisotopes in plankton change with time after fallout; initially the isotopes are in approximately the same ratio as they occurred in sea water but later specific **nuclides** are concentrated. From this evidence it would appear that the accumulation of radioisotopes shortly after fallout is principally by adsorption and later, by absorption.

Radioisotopes of manganese, iron, cobalt and zinc contributed 62 per cent of the total radioactivity in the plankton and almost 100 per cent in the fishes. Although the physiological role of these elements in fish is not known, it is interesting to note that in higher animals one or more of these elements are associated with oxygen and electron transfer, enzyme function and the structure of vitamin B_{12}.

Lowman also calculated the concentration of radioisotopes in omnivorous and carnivorous fishes as related to the radioelement content of the food upon which the fish were feeding. As these were field rather than laboratory studies it was necessary to make some assumptions about the fish-food relationship for specific individuals. From these studies it is concluded that omnivorous fish, which feed on plankton, almost completely exclude the fission products and, on a comparative basis, concentrate zinc and iron but discriminate against cobalt. Carnivorous tunas, which feed primarily on omnivorous fishes, discriminate in favor of zinc and manganese and against iron and cobalt, by comparison, to the percentage of these radioelements in their food supply.

Rice (6) discussed in more detail the relationship of phytoplankton to uptake as indicated in the title of his paper, "The Role of Phytoplankton in the Cycling of Radionuclides in the Marine Environment." Phytoplankton, which are one-celled plants living in the surface waters and are the foundation of the food pyramid in the ocean, accumulate a large number of elements from sea water and therefore, potentially, are an important means of transferring radionuclides from water to the food web.

However, the concentration of radionuclides by phytoplankton varies from species to species and therefore all species will not be equally important in the cycling of radionuclides in the cycling of radionuclides in the ocean. Phytoplankton accumulate radioisotopes from water rapidly but also may lose radioisotopes fairly rapidly in nonradioactive water by "biological dilution," the reproductive process of dividing one cell into two and thereby reducing by 50% the radioactivity in the original cell.

The transfer of radioisotopes from phytoplankton to a higher trophic level via the food chain and directly from the water also was investigated by Rice. He found that the concentration of radiocobalt by brine shrimp was 7.5 times greater when these animals fed upon radioactive phytoplankton than when water was the source of the radioactivity.

The effectiveness of phytoplankton in transferring radioisotopes from water to the food web also depends upon the cell size of the phytoplankton and the concentration of the cells in the water, because these factors largely determine if phytoplankton are acceptable food items for filter-feeding animals. Furthermore, the quantity of radioisotopes that are ingested by phytoplankton feeders is not an index of the amount of uptake since all species of phytoplankton are not digested with the same efficiency.

The paper by Corcoran and Kimball (7), "The Uptake, Accumulation and Exchange of Strontium-90 by Open Sea Phytoplankton" is a report of work of the same general nature as some earlier work by Rice, but in these experiments open sea species of phytoplankton, rather than inshore or fresh-water species, have been used. Another difference is than an estimate of adsorption was made by using heat-killed phytoplankton cells in the control lots. It was found that both Sr^{90} and Y^{90} were accumulated by the five stock cultures of oceanic phytoplankton used in this study but that the accumulation of Sr^{90} was apparently an adsorption process, since the accumulation in the heat-killed cells was equal to that in the living cells.

The paper by Pomeroy (8), "Experimental Studies of the Turnover of Phosphorous in Aquatic Environments" is not a report about a fallout nuclide but is of interest in that some methods for the study of phosphorous in an aquatic environment may be applicable to other elements.

The experiment was designed to evaluate the relative importance of biological and physical processes in the short term turnover of phosphorous in natural waters. The approach was to use an agent that would block a biological process and then observe the effect upon the turnover of phosphorous. For example, if the phosphate that is in rapid flux between water and plankton, bacteria and inorganic solids is being passed through some metabolic process, it should be possible to stop the rapid turnover of the phosphate by stopping key processes.

Agents used were methylene blue to block phosphorylation, cyanide to block respiratory metabolism and antibiotics to inhibit bacterial growth. For one part of the experiment aquaria were placed in the dark to stop photosynthesis. As a result of the addition of methylene blue and cyanide the short-term turnover of phosphate between the water and suspended solids was stopped, and therefore the short-term turnover is thought to involve biological rather than physical processes. Additions of antibiotics had variable effects which may be related to the size and character of the bacterial flora. In darkness the rate of turnover of phosphate is sometimes reduced.

A unique opportunity to observe the cycling of fallout radionuclides in a natural environment is available in the Marshall Islands at Rongelap Atoll, 80 miles east of Bikini Atoll. Rongelap was on the edge of the fallout pattern from the detonation of the 15 MT Bravo device at Bikini on March 1, 1954. The amount of fallout varied by a factor of ten between the northern and the southern islands of the atoll as indicated by the

gamma dose rates one day after the Bravo shot, which were 35 r/hr. and 3.5 r/hr. respectively. Fallout at Rongelap from other Bikini-Eniwetok detonations has been less than one per cent of the fallout which resulted from the Bravo detonation. Hence at Rongelap it is possible to observe the cycling of fallout **nuclides** in a natural environment, populated by man, and at various levels of radioactivity, all of which are safe for living but great enough to be easily measured. Furthermore, the fallout radionuclides at Rongelap essentially are of the same date of origin, March 1, 1954. To determine the fate of fallout radionuclides at Rongelap an ecological study was instituted in March 1958.

The distribution of radioisotopes as it existed at the atoll in the fall of 1959 is reported in the paper by Held (9), "Qualitative Distribution of Radionuclides at Rongelap Atoll." He found that the distribution of radionuclides in the marine and the terrestrial environments was distinctly different.

Approximately five years after the Bravo event the neutron induced radionuclides Zn^{65}, Co^{60} and Mn^{54} were the principal radionuclides found in marine organisms, while the long-lived fission products Cs^{137} and Sr^{90} were found primarily in the land organisms. All of the radionuclides found in lagoon bottom materials, marine plants, marine animals, soil, soil solution, land plants, land animals, and man are given on a distribution chart that indicates the relationship of the component parts to each other.

From the chart it is noted that Ce^{144} is the principal isotope in the marine algae and that corals and coralline algae contain some Sr^{90}. Twelve radioisotopes were detectable in soil, but Sr^{90}, Cs^{137} and Sb^{125} were the principal nuclides found in soil solution. The radioactivity in the land plants in general is 90 per cent or more Cs^{137} and the remainder Sr^{90}. This is unlike the situation usually found for continental soils and is a consequence of the low potassium content of Rongelap soil. Amendments of potassium to Rongelap soil reduce the uptake of Cs^{137} by plants and also affect the distribution of Cs^{137} within the plant.

The radionuclides in man at Rongelap are principally Zn^{65}, Cs^{137}, and Sr^{90}, which reflects a diet of both marine and terrestrial origin.

Two of the papers presented discussed the radioactivity in marine organisms collected in Connecticut in the vicinity of nuclear installations. The papers were, "Gross Beta Radioactivity in Marine Organisms" by Hatfield et al. (10) and "Zinc-65 in Oysters in Fishers Island Sound and Its Estuaries" by Fitzgerald and Skauen (11).

Hatfield et al. (10) observed seasonal variation in the gross beta activity of oysters, clams, mussels and algae which in general corresponded with the expected seasonal variation in metabolic activity. Values for all samples ranged from 17 to 438 $\mu\mu$c/g of ash; the highest values were for an alga, Laminaria (a kelp). The authors believe that naturally occurring K^{40} probably accounted for most of the beta activity noted. Further studies are in progress to determine if other beta-emitting **nuclides** are present.

Fitzgerald and Skauen (11) determined both Zn^{65} and stable zinc in oysters collected in the Thames River above and below the submarine base at New London, Connecticut. Stable zinc in moist tissue averaged about 0.2 per cent. The Zn^{65} values increased markedly during June and July, 1961, to a maximum of 11,500 μμc/kg (wet), which was six times greater than any previous value. The fact that it is unlikely that this increase was a consequence of fallout points to the need, when making surveys of fallout nuclides in the environment, to recognize that there may be sources of radionuclides other than fallout. The authors also determined the effect of temperature, zinc concentration, the presence of certain anions and of complexing upon the uptake of zinc. It was found that the uptake was slightly greater at 18°C. than at 5°C. but that the zinc concentration in the water had a greater effect upon uptake than temperature. A large amount of zinc was adsorbed on the shell except when the zinc was chemically complexed. The uptake of zinc from zinc chloride, sulfate, phosphate and tartrate was similar for all four compounds and reached a maximum within 24 hours.

The final paper to be reviewed, "Micro-organism Behavior in Radioactive Environments" by Lackey and Bennett (12) is not directly related to fallout. It is a report of a search for a biological indicator of ionizing radiation based upon the observed response of microorganisms to a radiation field. There are many environmental factors which an organism can sense and react to, but as a result of field observations and laboratory experiments by Lackey and Bennett, ionizing radiation apparently is not one of these factors.

The assemblage of organisms in areas of seepage from high level waste pits and in a sedimentation pond at Oak Ridge was compared with the assemblage of organisms elsewhere in the Oak Ridge area where similar soil conditions prevailed but without radioactivity. With one exception the assemblages were the same and from these observations it was concluded that the field occurrence of microorganisms in a radioactive environment is not different from that of a similar, but nonradioactive, environment.

The authors have had several years experience of exposing microorganisms to ionizing radiation in various sorts of laboratory experiments but have observed no visible response to alpha, beta or gamma radiation, other than death. Although the information about radiation exposure was limited, one experiment was cited in which the LD_{50} for marine bacteria exposed to a Co^{60} source was 1,500,000 r and the dose for total kill was 3,000,000 r. In another experiment, the genetic damage of an LD_{50} dose to a protozoan (Entosiphon zulcatus, a colorless euglenid some 30 μ in length) was measured by observing the daily division rate of the survivors. In this case, the daily division rate of the survivors was followed for 60 generations and no effects of the irradiation were detected. Other parts of the experiment demonstrated the preferential uptake of certain radionuclides by some organisms. In conclusion Lackey and Bennett state that no change, morphological, physiological, or behavioral, of the microorganisms was observed, using gamma or alpha or beta sources of irradiation.

Papers Reviewed

1. Bowen, V. T., and T. T. Sugihara. Cycling and Levels of Strontium-90, Cerium-144, and Promethium-147 in the Atlantic Ocean. Woods Hole Oceanographic Institution, and Clark University. Symposium Paper* No. 19.

2. Duke, T. W., E. R. Ibert, and K. M. Rae. Availability of Sediment-sorbed Materials to Marine Biota. Agricultural and Mechanical College of Texas. Symposium Paper No. 26.**

3. Seymour, A. H. Radioactivity of Marine Organisms from Guam, Palau and the Gulf of Siam, 1958-1959. University of Washington. Symposium Paper No. 22.

4. Townsley, S. J., D. F. Reid, and W. T. Ego. The Effect of Environmental Ions on the Concentration of Radiocalcium and Radiostrontium by Euryhaline Teleosts. University of Hawaii, Symposium Paper No. 35.

5. Lowman, F. G. Radionuclides in Plankton and Tuna from the Central Pacific. University of Washington. Symposium Paper No. 21.

6. Rice, T. R. The Role of Phytoplankton in the Cycling of Radionuclides in the Marine Environment. Bureau of Commercial Fisheries, U. S. Fish and Wildlife Service, U. S. Department of Interior, Beaufort, N. C. Symposium Paper No. 33.

7. Corcoran, E. F., and J. F. Kimball. The Uptake, Accumulation and Exchange of Strontium-90 by Open Sea Phytoplankton. University of Miami. Symposium Paper No. 34.

8. Pomeroy, L. R. Experimental Studies of the Turnover of Phosphorus in Aquatic Environments. University of Georgia. Symposium Paper No. 24.

9. Held, E. E. Qualitative Distribution of Radionuclides at Rongelap Atoll. University of Washington. Symposium Paper No. 25.

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* Papers presented in Session II: Section 2, Part A, Cycling and Levels of Nuclides in the Marine Environment, of the First Symposium on Radioecology, Colorado State University, September 10-15, 1961. (Proceedings to be published, U. S. Government Printing Office, Washington, V. Schultz and A. W. Klement, Jr., eds., 1962).

**Not presented during the symposium.

10. Hatfield, T. W., D. M. Skauen, and J. S. Rankin, Jr. Gross Beta Radioactivity in Marine Organisms. University of Connecticut. Symposium Paper No. 20.

11. Fitzgerald, B. W., and D. M. Skauen. Zinc-65 in Oysters in Fishers Island Sound and its Estuaries. University of Connecticut. Symposium Paper No. 23.

12. Lackey, J. B., and C. F. Bennett. Micro-Organism Behavior in Radioactive Environments. University of Florida, and Scripps Institution of Oceanography, University of California. Symposium Paper No. 28.

RADIONUCLIDES IN THE FRESHWATER ENVIRONMENT*

Louis A. Krumholz
Department of Biology
University of Louisville
Louisville, Kentucky

Within the past fifteen years or so, much has been written about the accumulation of radioactive nuclides by aquatic organisms. Those reports are concerned primarily with the uptake, accumulation, retention, and/or loss of radioactive materials and their cycling through the environment. However, a great deal of that work has been confined to the assessment of effects of radioactive waste materials in particular areas, especially in the vicinity of atomic energy installations, or to the study of the effects of known concentrations of particular radionuclides on various organisms or communities.

Most studies concerned with radioactive fallout materials have been performed in marine waters, and there have been relatively few studies in the freshwater environment. The opportunities for studying the effects of fallout over the ocean were greatly enhanced by the series of tests at the Eniwetok Proving Ground, and much has been learned about the dispersal and cycling of those nuclides through the oceans. To the contrary, there have been no similar tests in the vicinity of large bodies of fresh water, and, as a result, there is little information available of fallout in freshwater communities.

There are several reasons why much of our research has been confined to intensive studies of particular radionuclides with certain selected organisms:

1. The relative ease of control in handling particular radionuclides,

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either singly or in combination, in known tracer amounts.

2. The relative ease in interpreting the data when only one or two nuclides are under consideration.

3. The elimination of many environmental factors by using selected organisms under more or less controlled conditions.

4. The expressed need for immediate information on those nuclides which are long-lived and of possible harm to man.

At the same time, there are reasons for not undertaking extensive studies of the cycling of the complete spectrum of radioactive fallout materials through natural ecosystems:

1. The difficulty in assaying with accuracy the amounts and identities of fission elements in quantities at acceptable fallout levels, especially in regions relatively remote from nuclear detonations.

2. The general unavailability, until recently, of instrumentation sufficiently sensitive to detect and measure accurately the nuclides in various parts of the biota and environment. Such instrumentation usually has been available only at special laboratories.

3. The assumed danger of releasing known tracer amounts of fission products, especially the long-lived radionuclides of strontium, cesium, and some of the rare earth metals, into any environment over which strict control could not be maintained.

4. The scarcity of adequately trained personnel, capable of and having the time required to oversee and perform such studies.

In the recent Symposium on Radioecology at Colorado State University, various aspects of the cycling and levels of radionuclides in the freshwater environment were discussed. Three papers (1,2,3) were concerned with waste materials from major atomic energy installations, and one (4) with the cycling of radiophosphorus in a stream ecosystem, and one (5) with the occurrence of radium in natural waters, and one (6) with the cycling of natural radioactivity, including fallout materials, in a stream ecosystem. (These papers are listed at the end of this review as is the other literature cited).

Nelson (1), in his study of strontium and calcium relationships in mollusks from the Clinch and Tennessee Rivers, found that all clams accumulated both stable and radioactive strontium in their shells. Nelson considered the low levels of Sr^{90} released from the Oak Ridge National Laboratory through White Oak Creek as tracer amounts, and used that nuclide to determine the cycling of strontium through mollusk shells. Clams collected from areas upstream from the mouth of White Oak Creek contained considerably smaller amounts of Sr^{90} than those taken downstream from the mouth of the creek.

The amounts of Sr^{90} accumulated by the clams below White Oak Creek decreased progressively the farther they were downstream, and there was close correlation between the stable/radioactive strontium ratios observed in the waters where collections were made and those expected on the basis of dilution of Clinch River water by the Tennessee River.

The rate of shell deposition and the strontium content of the shells of Anodonta corpulenta increased with the age of the animals, and in that species the strontium deposition was directly proportional to the growth rate and inversely proportional to the surface/volume ratio. It is apparent from his data that the clams were unable to distinguish between the radioactive and stable nuclides of strontium. In Pleurobema cordatum, shells from clams of the same age, but taken from localities about 187 miles apart, had respective growth rates of 2.5 and 4.9 g/yr. and carried 201 and 237 μg Sr/g of shell, respectively. Those specimens with the slower growth rate and the lower amounts of strontium were taken from the Clinch River about 26 miles above the mouth of White Oak Creek, whereas the others were collected from the Tennessee River about 160 miles downstream from the mouth of White Oak Creek. In that instance, it is likely that the lower water temperatures in the Clinch River (just below Norris Dam) could have been a causative factor in the slower growth rate and the consequent lower accumulation. Also, in those samples he noted that the clams from the Clinch River (above White Oak Creek) contained only about 0.2 μμc Sr^{90}/g of shell, whereas those taken from the Tennessee River well below the creek mouth contained about 4 μμc Sr^{90}/g of shell. The highest accumulations of Sr^{90} reported in this study were 99 μμc/g of shell in Anodonta corpulenta and 15 μμc/g of shell in Quadrula pustulosa taken from the Clinch River; the highest accumulation in shells from the Tennessee River was about 25 μμc/g of shell in Elliptio crassidens collected about 45 miles below the mouth of the Clinch River.

Nelson stated that the difference in mean strontium content among species was due to inherent differences in the animal species rather than to any differences in the strontium/calcium ratios in the environment. In this respect, his findings were in contrast with those of Odum (7,8), Swan (9), and Thompson and Chow (10). It has been pointed out by Revelle and Schaefer (11) that the ratios of the more abundant elements in sea water are very nearly constant despite variations in absolute concentrations in different parts of the oceans. At the same time, Krumholz, Goldberg, and Boroughs (12) stated that whereas the oceans may be considered very roughly as a homogeneous mass, fresh waters have a tremendous range in their chemical characteristics. Thus, while the calcium/strontium ratio in ocean waters may be more or less constant, that ratio varies widely in fresh waters, and those environmental differences in the ratios could easily explain the discrepancies between the findings of Nelson and the other workers cited.

Hemphill, Nix, and Carrigan (2) working in the Clinch River below the Oak Ridge National Laboratory, found that there was an uneven distribution of radioactivity in bottom sediments. They took surface readings and made test borings of the sediments in transects across the river at various distances below the outfall of White Oak Creek, and found a good correlation

between the distribution of silt deposits on the bottom and the radioactivity. Also, there was a general gradual diminution in the amounts of radioactivity the farther downstream from the mouth of White Oak Creek the samples were taken.

When White Oak Lake was drained in 1955, most of the accumulated silt remained in the lake bed, but that portion of silt directly in the stream channel was washed out and carried into the Clinch River. That "slug" of relatively "hot" silt has gradually been moved downstream at a rate of about two miles a year.

Ophel (3) reported on the fate of Sr^{90} in a freshwater lake at the Chalk River establishment of Atomic Energy of Canada Limited. He used Sr^{90} that had seeped from a liquid disposal area into the inlet stream of Perch Lake as a tracer to follow the passage of that nuclide through the lake ecosystem. During the 5-year period, 1956 through 1960, a total of about 4000 mc Sr^{90} entered the lake and about 3000 mc left the lake during the same period. He showed, through periodic sampling, that there were seasonal fluctuations in the amounts of Sr^{90} in the lake water from winter to summer, the amounts in winter being consistently higher than those in summer. He attributed the depletion of radiostrontium during the warmer months of the year largely to the increased biological activity in the lake.

During the first two years after the radiostrontium entered the lake, the radioactive content of the water slowly increased and then remained fairly constant during the last three years. Meanwhile, the Sr^{90} content in the bones of adult yellow perch (Perca flavescens) gradually increased over a 4-year period, and finally reached an equilibrium in the fifth year. The content of Sr^{90} in the bones of adult perch represented a concentration factor of about 700 times that in the lake water in 1956 and 1957 as compared with one of about 3000 times that of the lake water when equilibrium was reached. These findings indicate that there is a gradual buildup of Sr^{90} in the bones of fishes over a relatively long time during which they are constantly exposed to that nuclide in the natural environment.

In various parts of the environment and the biota, Ophel reported concentration factors of 200 for the bottom sediments (the top inch), 300 for aquatic plants, 750 for clams, 1000 for minnows and bones of mink, 1400 for bones of beaver, and 3900 for bones of muskrat. Also, he theorized that the concentration of Sr^{90} would continue to increase in the bones of the mammals because parts of their lives had been spent in uncontaminated areas, and they had not yet reached equilibrium with the environment.

He concluded from his findings that, because most of the Sr^{90} was concentrated in the bones of animals, the major pathway of radioactive strontium from a contaminated body of water could be from direct use for drinking purposes, and not by way of food webs originating in the aquatic environment.

Scott (5) presented a resume of findings of the U. S. Geological Survey

in its survey of the occurrence of radium in natural waters of the United States. Principal emphasis was placed on samples of water taken from deep wells from the north central to the southwestern and Rocky Mountain States.

Ball and Hooper (4) described a thorough study of the movement of radiophosphorus through a cold-water stream ecosystem in northern Michigan during the summers of 1958, 1959, and 1960. They added 23 P^{32} to the West Branch of the Sturgeon River at a uniform rate so as to fix the radioactivity in the producer level of the food web within a short span of time. The addition of radiophosphorus was repeated each year at the same location. Samples of water and of the biota were collected periodically for from 5 to 7 weeks each year; the organisms sampled included the periphyton, aquatic macrophytes, invertebrates, and fishes, representing the various trophic levels.

They found that the periphyton, the film of microscopic plants (principally diatoms) that covered the rocks and other substrates in the stream, was the most important single source of biological uptake as indicated by the activity-density curves. However, the data were variable principally because of differences in location, current, shading, growth rate, species composition of the samples, thickness of film, and so forth. The pattern of uptake of radiophosphorus by periphyton was quite different at different stations each year; the greatest uptake at all stations occurred in 1960, the least in 1959, and in 1958 the uptake was intermediate. Their impression was that most of the P^{32} in the periphyton was intercellular.

The principal aquatic macrophytes in the stream were the alga, Chara sp.; the moss, Fontinalis antipyretica; and the vascular plants, Potamogeton pectinatus, Nasturtium officinale, Elodea canadensis, and Ranunculus sp. Of these, only the first three were abundant. The activity-density curves for Chara and Potamogeton were similar, but those for the moss were consistently at a higher equilibrium level, in most instances 2 or 3 times greater. An interesting feature of the activity-density curves of Fontinalis and Nasturtium was that they bimodal, showing a secondary rise in the late summer of 1960 that probably resulted from uptake following the decay of detritus in the bottom sediments. In no instance did the levels of radioactivity in the macrophytes reach those attained in the periphyton. However, the year-to-year changes in uptake by the macrophytes was opposite to that of the periphyton; the maximum uptake by macrophytes occurred in 1959 and the lowest uptake was recorded in 1960.

The studies on invertebrates did not include all species found in the stream, but rather were confined to those found in sufficient numbers to provide an adequate weekly sample and to those that remained in the study area throughout the period of observation. The principal animals for which analyses were made included various insect larvae such as those of the snipe fly, Atherix; the caddisflies, Brachycentrus and Hydropsyche; the stonefly, Pteronarcys; the mayflies, Hexagenia limbata, Ephemerella cornuta, and E. needhami; the blackfly, Simulium; and the fishfly,

Nigronia. Unfortunately, most of these invertebrates were not available in large enough numbers to have been sampled each of the 3 years; the only one was the blackfly larva. Some of the insect larvae contained more radioactivity one year than another, and no insect was consistently higher in radioactivity every year.

Ball and Hooper found that the activity-density curves of the insect larvae suggested the following phenomena:

1. The similarity in the shape of the curve for each station each year for a given species indicated that the curve was characteristic of the animal rather than the environment.

2. There was a wide range in maxima for a given species that differed greatly from year to year, and appears to be closely related to stream conditions at the time the observations were made.

3. Curves were similar for closely related species living in different microhabitats.

4. Smaller forms showed consistently higher levels of radioactivity than larger forms.

5. Many of the differences in the slopes of the curves could not be explained on the basis of animal size alone.

6. In many instances radioactivity was being lost in one species while being taken up at an increasing rate by another species.

The only fishes studied intensively were the sculpin, Cottus cognatus, and the brown trout, Salmo trutta. These animals usually retained radioactivity longer than many of the invertebrates, and the uptake was about the same for both species both years they were taken (1958 and 1959).

The broad trophic levels could be quantitatively distinguished from one another by careful study of the activity-density curves, and the uptake of radiophosphorus could readily be traced on a basis of time through the producers, the primary consumers such as the filter feeders, periphyton scrapers, omnivores, and detritus feeders, and ultimately to the carnivores as the secondary consumers.

Minckley, Craddock, and Krumholz (6) gave a preliminary report on the passage of naturally occurring radionuclides through the food web of the banded sculpin, Cottus carolinae, in a torrent spring near Louisville, Kentucky. Bi-weekly collections of physical, chemical, and biological samples from Doe Run were made from 2 stations in the stream for a 2-year period (1959-1961). The kinds of organisms comprising the food web of the sculpin were based on food preferences indicated by analyses of stomach contents of fishes of different sizes at different times of the year. The food preferences at the 2 stations were quite different, and

it was obvious that the sculpins preyed upon almost any animal they could swallow. There was a marked change in food habits as the fish grew larger. At Station I, the small sculpins fed principally on chironomid and other insect larvae, amphipods, and isopods, but as they grew larger their diet included an increasing number of decapod crustaceans. At Station IV, where there were fewer amphipods and more chironomid larvae, the diet of the small fish was principally chironomids and to a lesser extent the larvae of mayflies. As those fish grew, the diet included more mayfly larvae, and at an even greater size, the sculpins finally ate more and more crayfishes and fishes. At both stations it was apparent that the principal time for feeding was in the early part of the night (from 6:00 P.M. until midnight).

Analyses of the stomach contents also provided information on the utilization of foods eaten; that is, the amounts of foods eaten that were transferred into protoplasm. The utilization of food ranged from 5.2 to 14.3% of the food consumed when compared with the weight of the fish, but was fairly uniform for fish of all sizes. These data were supplemented by experiments which showed that the time required for the digestion and elimination of food materials at the temperatures prevailing in Doe Run was about 36 hours.

Samples of the biota from each trophic level were assayed for gross beta radioactivity, and the different contributing radionuclides were determined by gamma-ray spectrometry or radiochemical analysis. The principal naturally occurring radionuclides in the stream ecosystem were daughter products of U^{238} and Th^{232}, measured as Bi^{214} and Ac^{228}, respectively. The principal fallout materials were Sr^{90} and Cs^{137}, although some Zr^{95}, Ru^{106} and Ce^{144} were reported.

During the early part of the study and during the first 8 months of 1961, the amounts of fallout materials gradually decreased until by August 1961, the naturally occurring radionuclides made up much the greater part of the total radioactivity in the environment.

Since September 1961 there apparently has been a small but steady increase in accumulation of radionuclides, but the data are too preliminary to warrant critical examination. The average amount of radioactivity carried by the water of Doe Run, based on 49 1-liter samples, was about 10μμc/l. The marl substrate of the stream contained about 4μμc/g of material. As is to be expected, some radioactivity was accumulated by the different organisms in the food web of the sculpin. The algae, with the exception of the red alga, Batrachospermum moniliforme, contained from 8 to 15μμc/g wet weight; 12 samples of the red alga contained an average of only 1.5 μμc/g. The moss, Fissidens julianus, contained an average of 8.8μμc/g for 31 samples. Among the consumer organisms for which radiochemical or spectrometric analyses are available, there is an indication of a marked elemental selectivity of the radionuclides present. It has not been demonstrated that the naturally occurring radionuclides in Doe Run are utilized in the metabolic processes of plants and animals,

but there is ample evidence that the radionuclides produced by nuclear fission are. Accordingly, an accumulation of radioactive nuclides from nuclear fallout might be expected to be acquired by organisms living in their natural environment, whereas those of the naturally occurring transuranic elements might not.

Among the primary consumers, such as the isopods, amphipods, snails, and insect larvae, there was a much wider range in accumulation of radioactive materials within each species than among the producers. In almost every instance there were samples of organisms that did not contain measurable amounts of radioactivity while others contained relatively large amounts. Such a wide range in accumulation is not peculiar to the animals in Doe Run, but has been noted by many other workers. Of those animals collected at both stations, the isopods contained greater average amounts and showed greater ranges of uptake than any other organism; at Station I the average accumulation was 23.3 μμc /g wet weight, with a maximum of 104 μμc /g, and at Station IV the average was 22.9 μμc /g with a maximum of 92. Next highest in accumulation were the caddisfly larvae with averages at Stations I and IV of 17.9 and 16.6 μμc /g, respectively, and with respective maxima of 67 and 31. Radiochemical analyses of those samples indicated that about 10% of the total radioactivity was traceable to Sr^{90} and Cs^{137}.

Of those primary consumers taken only at Station IV, the highest reported radioactivity was that of stonefly larvae that contained 156 μμc /g; the average of 26 samples was 20.7 μμc /g. The next highest accumulation was that for mayfly larvae with a maximum of 105 μμc /g and an average of 17.6 μμc /g for 38 samples. Radiochemical analyses of those samples showed that 27% of the radioactivity in the stoneflies was traceable to Sr^{90} and Cs^{137}, whereas in the mayflies, 24% of the radioactivity was from those two radionuclides.

The snail, Goniobasis, which feeds primarily on the encrusting periphyton that contains relatively great amounts of radioactivity, accumulated relatively little radioactivity. In 31 samples from Station I, the average accumulation was 2.0 μμc /g with a range of 0 to 6 μμc/g, whereas in 29 samples from Station IV, the average accumulation was 4.0 μμc /g with a range of 0 to 15 μμc /g.

Among the crayfishes, of which Orconectes rusticus is at best a primary consumer and Cambarus bartoni is at times a primary, a secondary, or even a tertiary consumer, it is obvious that Orconectes accumulated considerably greater amounts of radioactivity. The average for 13 samples of Orconectes was 3.9 μμc /g of which 11% was traceable to Sr^{90} and Cs^{137}, and the average of 14 samples of Cambarus was 1.2 μμc /g of which about 8% was from Sr^{90} and Cs^{137}.

Among the fishes, the average amounts of radioactivity were about the same as for the crayfishes. For the minnow Notropis spilopterus, the average for 8 individuals was 1.8 μμc /g with a range from 1 to 5 μμc /g.

For the darter Etheostoma flabellare, the average of 15 samples was 3.0 μμc/g with a range from 0 to 5 μμc/g. Both of these species were taken only from Station IV. In samples of the sculpin, which were taken from both stations, the average for 9 individuals from Station I was 1.2 μμc/g of which about 9% was contributed by Sr^{90} and Cs^{137}, whereas for 6 individuals from Station IV, the average was 1.0 μμc/g of which 43% of the radioactivity was from Sr^{90} and Cs^{137}.

Based on our over-all data, we found that there was a decrease in the amount of radioactivity per gram of food ingested by sculpins in Doe Run as those fish increased in size. However, the rate of that decrease did not correspond with the rate of decrease in the radioactivity of the fish themselves, and we believe that an actual change in metabolism is as important in determining the accumulation of radioactivity as are the amounts of radionuclides ingested. From the data, we calculated that the percentage of ingested radioactivity retained in the body of the sculpin is relatively high early in life (6.5%), but that just before maturity is reached that percentage drops to about 1.5% and then increases very slowly throughout life but does not exceed about 2.5%.

Davis, Hanson, and Watson (13) in speaking of the effects of environmental factors on the accumulation of fallout on natural populations, stressed the differences in the sorption of fallout materials as influenced by the amount of rainfall available for dilution. Although Pendleton and Uhler (14) showed experimentally that there was a direct relationship between the uptake of Cs^{137} and the amount of water in the system, Davis and his coworkers showed that the relationship does not persist as the amount of water continues to increase. They compared the amount of fallout radionuclides in plants from wet tundra in arctic Alaska with emergent aquatic plants from a shallow pond in the same region, and attributed the lower levels of radionuclides in the aquatic plants to the dilution of fallout by the pond water and to the removal by plankton organisms. Also, they showed that in two species of birds, the ptarmigan which inhabits and feeds in the tundra and the oldsquaw duck which feeds in the pond, the ptarmigan consistently accumulated greater amounts of fallout radionuclides than did the duck. There, they said that the lower levels of radioactivity in the ducks demonstrated the importance of radionuclide content in the primary producers on the amounts accumulated in organisms of the higher trophic levels.

Those same men showed that water drained from a barren, high-altitude watershed in Washington State carried the radioactive fallout material to a small lake, and that the moss living at the brim of a waterfall at the overflow of the lake essentially functioned as a filter for the radionuclides in the water, and as a result contained from 4 to 11 times as much radioactivity as moss collected from an exposed site in a nearby rain forest.

By contrast, Osburn (15) reported that in the alpine tundra of Colorado the fallout materials brought down by the snowfall were essentially

filtered out as the melt water passed through the accumulated organic detritus in the region, and was virtually free from radioactive contamination by the time it reached the streams. Osburn, too, found that the mosses in the tundra were excellent filters for radioactive fallout materials.

In another study* conducted at the University of Louisville, under the sponsorship of the Ohio River Valley Water Sanitation Commission (ORSANCO) (see papers listed as Anonymous (16)) an effort was made to determine the amounts of radioactive fallout accumulated in various parts of the ecosystem of the Ohio River. Samples of water and different organisms from each trophic level in the stream were collected at various times of the year and assayed for gross beta radioactivity. The amounts of radioactivity in samples of water from the river near Louisville gradually declined from about 75 to 85 μμc/l in the summer of 1958 to from 9 to 80 μμc /l in 1959. At the same time, the levels of gross beta radioactivity in a 70-acre lake in southern Indiana near Louisville gradually increased from an average of about 120 μμc /l in 1958 to an average of nearly 135 μμc/l by late 1959. These preliminary data indicate that as the total amount of worldwide fallout gradually decreases, the stream ecosystem gradually cleans itself of the accumulated radioactive materials, whereas those long-lived radionuclides impounded in a more or less stationary body of water will remain there for the rest of their physical half lives. Also, as succeeding rains wash in more and more fallout, even though the total amount may be decreasing each year, there will be a gradual buildup of those long-lived fission products in the impounded area. Such a condition probably occurs throughout the world, and would be especially evident in areas where small impoundments are used as water supplies for livestock. Most such ponds are constructed so that nearly all the water from the watershed will be contained within the pond, although provisions are made for an emergency spillway to take care of intermittent flooding.

Another series of data that corroborates these findings concerns the fishes in the two environments; the radioactivity in 3 small bluegills (Lepomis macrochirus) from the Ohio River amounted to about 7 μμc /g for the whole fish, while for 6 bluegills from the lake the average accumulation amounted to about 23 μμc/g.

Samples of organisms taken from the Ohio River during this ORSANCO-University of Louisville survey indicated that the filamentous algae accumulated gross beta radioactivity up to about 90 μμc/g, and that net plankton taken in tows with a 20-mesh silk bolting cloth net contained about 40 μμc/g wet weight. Live clams accumulated up to 30 μμc/g of muscle, and live snails up to about 40 μμc/g. Apparently the snails were quite selective in their metabolism of radionuclides since their excrement contained up to about 200 μμc/g. The shells of dead clams and snails picked up along the river bank did not contain measurable amounts of radioactivity.

*This project is under the direction of Dr. William M. Clay

Among 11 species of fishes sampled, the amounts of gross beta radioactivity ranged from 0 to 20 μμc/g of whole fish, and there was a marked range of accumulation for individuals within each species sampled. Using two common food fishes, the channel catfish (Ictalurus punctatus) and the freshwater drum (Aplodinotus grunniens), as examples, there was a marked loss in radioactivity for fishes of the same size range from 1958 to 1960. The radioactivity in 17 individual catfish ranged from 3.3 to 7.3 μμc/g, and in 36 individual drum it ranged from 1.3 to 10.9 μμc/g, with respective averages of 5.3 and 4.5 μμc/g. By 1960, in 51 catfish collected and assayed during the first half of the year, the range had fallen to from 0.7 to 3.0, with an average of 1.5 μμc/g, and for 136 drum the range was from 0 to 4.2 with an average of 1.5 μμc/g. All assays were made on individual whole fish that ranged in weight from 3 to 15 g. From these data it is apparent that there is a direct correlation between the amount of radioactivity in the river water and that of the fish living in the water; the average radioactivity in the water in 1958 was about 80 μμc/l and in 1960 was about 32, whereas in the catfish and drum the average radioactivity was 5.3 and 4.5 μμc/g, respectively, in 1958, and in 1960 was 1.5 for each species.

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DISCUSSION

Mr. Alfred W. Klement, Jr., Chairman of the session, directed the discussion of the three review papers presented.

KLEMENT: While these have been summaries of a number of papers themselves, a great deal of the work in these areas has been done by the individuals here, including a number of people in the audience. I think among the group here we can discuss these subjects in some detail.

HELD: Dr. AUERBACH, to what do you attribute the reduction of radioactivity in plants after burning of an area?

AUERBACH: I think the greater accumulation on unburned grass was due to residual contamination on the foliage, and burning the plants simply releases the ions, and the materials are fixed in the soil surface and are either not as leachable or as available as the Ca, the K, and the other elements in the plant materials.

GLASS: You suggested controlled burning as a possibility. What is likely to happen to the radionuclides then? It reminds me of the person who tried to get rid of poison ivy by burning it and had one of the worst attacks I have ever seen, because of the effects of the smoke.

AUERBACH: This would depend on the particular nuclides. Elements such as Cs are not very volatile under 450° C, nor is Sr very volatile under this temperature. And I think in most grass burning you would not achieve those temperatures. The application I had in mind was really for large installations. Rather than attempt to harvest and remove the crops to reduce a localized contamination problem, it might be equally satisfactory to use the burning method.

GLASS: It is transferred to the soil, then, isn't it?

AUERBACH: Yes.

RUSSELL: Have any measurements been made of the Pu content of fresh waters and their sedimentation rates?

KRUMHOLZ: I don't know of any, perhaps Jerry DAVIS knows of some.

Jared J. DAVIS: (Hanford Laboratories, General Electric Co., Richland, Washington). I know of no such measurements.

GRUMMITT: We have made some measurements on the Ottawa River, in shallow wells and local lake at Chalk River. The levels are so low as to be almost immeasurable, ranging from 2×10^{-3} to 10^{-2} $\mu\mu c/l$.

DAVIS: Earlier this morning there arose the question by Langham of the reindeer among the Laplanders. We have been conducting some studies that are quite comparable to this in the Artic with caribou, which are the same as the reindeer. We are finding levels of Cs in caribou quite comparable to those found in Lapland reindeer. The same report referred to lichens also as a source of Cs to the reindeer. We, also, are sampling lichens. Lichens do have about the highest concentration of Cs of any of the plants we have sampled in the Artic. However, I shot a caribou in a lichen community, and we failed to find any lichen in its gut content. Its gut content seemed to contain largely sedges, willow, and other miscellaneous plants from the tundra. Generally we find the highest levels of radionuclides in the vegetation from marshy areas. This would help explain the higher level in reindeer, or caribou, I think. Now, animals such as the ground squirrels will have significantly lower radioactivity in the flesh than will the flesh of reindeer on the order of 4 to 20 times lower.

I was interested in the discussions about the levels of radioactivity. If we grossly compare the levels of radioactivity in vegetation from comparable communities in the Arctic versus comparable rainfall areas in Washington (State), we find that the levels are a little higher, in the Arctic on the order of about twice as high.

KRUMHOLZ: There seems to be a contradiction between what you just said and the statement I referred to in your paper in which you said the amount of radioactivity in the aquatic plants was lower than in the terrestrial plants. Here you say, the uptake was higher in aquatic plants.

DAVIS: There are exceptions to the rules, but the highest levels usually occur in plants from marshy areas, as mentioned this morning where it is moist. If the water becomes too deep, there is a dilution factor that apparently takes over. So if you take soil of field capacity and increase the amount of moisture from moist soil to a marshy area, there is a marked increase in radioactivity, even in the same species of plant. However, in a truly aquatic situation, for example, in a point 8dm deep, there will be a very definite decrease. The lowest we have found occurred in emergent plants that did live in ponds. A deep pond would have a very, very low concentration of radioactivity in its biota. There are also plants in dry habitats that have high levels, and lichen is one good example. This is due, of course, to its peculiar habits. It is perennial, living there for years and years, and is hygroscopic. Such special properties make the picture a little different.

With some of the laboratory experiments at Hanford, we find if you go into a very dry situation, the accumulation of Cs is amplified again. So we know that under very dry conditions there may be an increased uptake.

DISTRIBUTION AND CYCLING OF FALLOUT NUCLIDES

PART 5. FALLOUT NUCLIDES IN THE FOOD CHAIN AND IN MAN - REVIEWS

Session Chairman: S. Allan Lough
Assistant Director for
Radiological Physics
Division of Biology and Medicine
U. S. Atomic Energy Commission

STRONTIUM AND BARIUM IN BONE AND DIET

William E. Grummitt
Chalk River Project
Atomic Energy of Canada Limited
Chalk River, Ontario, Canada

INTRODUCTION

One of the problems associated with the prediction of the effects of fallout from nuclear weapons is the calculation of levels of Sr^{90} in bone from known intakes in diet. Perhaps the easiest way of achieving this is by measuring the natural Sr content of the components of diet and relating this in turn to bone.

The results can be used either directly for calculating Sr^{90} specific activities and hence equilibrium bone levels, or alternatively, all measurements can be related to Ca in which case the bone level is calculated by means of the O.R. (observed ratio) bone/diet. In actual fact it makes little difference which method is used, as Ca data are available anyway and the result is identical in each case. However, the use of O.R.'s is slightly more complex and tends to confuse the layman. Measurement of stable Sr concentrations could possibly be avoided if a reliable value for the O.R. were available from an independent experiment. Such is not at present the case.

EXPERIMENTAL

In the present study, both Ba and Sr were measured by neutron activation analysis of the ashed material. Samples of produce were collected in as representative a manner as possible, frequent assistance being obtained from the Department of Agriculture. Activation analysis was chosen because:

(a) its reliability is good (estimated coefficient of variation $\pm$ 2%),

(b) it offered the possibility of doing Sr and Ba simultaneously, and
(c) samples differed widely in type and composition.

For this type of sample neutron activation was more rapid than flame photometry.

Results

Diet. Let us first of all consider Sr in diet, then for comparison look at the results for Ba. Some 30 components of diet have been analyzed for these two elements and for Ca. Items were selected on the basis of their relative importance in diet and samples were collected in several Canadian cities in order to give as representative a value as possible.

The results for Sr fall in a fairly narrow range, when expressed per gram of Ca. For instance: cereals, lugmes, nuts and vegetables lie in the range 1.4 to 8.2 mg/g Ca and animal products such as meat, milk and bone lie in the range 0.39 to 0.74 mg/g Ca.

The total daily intake is found to be 1.29 g of Sr and 0.40 g of Ba. In almost all instances we have underestimated the Sr content of food so that additional analyses have increased the apparent Sr intake.

Ba results show considerably more scatter from sample to sample than Sr, no matter what basis is chosen for comparison. Values ranging from 0.6 to 12 mg/g Ca (leafy vegetables to bran) are considerably higher than those for animal produce which range from 0.02 to 0.8 mg/g Ca. This is 5 to 10 times as great a spread as that observed in the Sr results.

Ba in the Canadian diet appears to be derived about 25% from milk, 25% from flour and up to 25% from potatoes. In contrast, milk is the only major contributor of Sr to the diet.

Distribution in Bone. In order to assess the value of bulk samples, we have studied the distribution of Sr in the population and in various components of the skeleton. A limited number of results have indicated that the skeletal distribution of Sr is nearly normal with minimum and maximum limits of only 0.7 and 1.3 times the mean. There appears to be no tendency to concentrate in a particular bone.

The population distribution, on the contrary, is rather skewed with some values up to 7 times the mean (Fig. 1). At the moment it is not known whether the second peak reflects a difference in composition of diet but the asymmetry is present whether one uses data from a single city (Montreal) or combines these to produce a composite diagram.

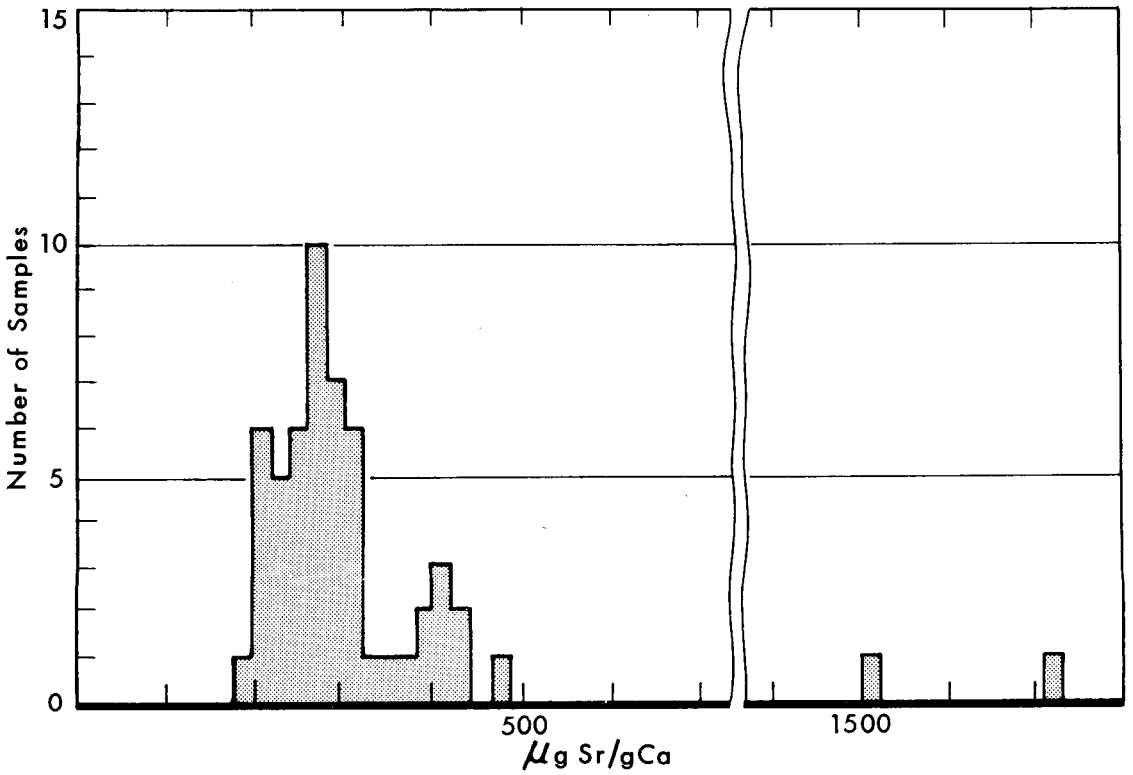

Figure 1. Distribution of natural Sr in the population, expressed per gram of Ca in bone. The histrogram depicts the number of samples which have a Sr/Ca ratio within a given concentration interval.

The results are quite different from those of Thurber and Kulp[1] for New York City, who report a normal distribution and values of only twice the mean.

At this point, some comment about high values might be desirable. The first of these was obtained some years ago and was substantiated by duplicate analyses. The second was similarly duplicated, but fortunately 3 other bones were available from the same skeleton. The analyses of these gave:

Vertebra	1530, 1509	μg/g Ca
Sternum	1570	" "
Rib	1360	" "
Long Bone	830	" "

For comparison, the median for the city of Montreal was about 280 μg/g Ca.

No age effect was observed in this study, the diet apparently being sufficiently constant with age to minimize such effects. Averages for the 0 to 19 and 20 to 70 year old groups are 300 and 320 μg/g Ca respectively.

The shape of the distribution curve for Ba is also skewed with values up to 5 times the mean. As one might expect from the diet study, the Ba results show more scatter than those for Sr, particularly for various bones within a single skeleton. Ba and Sr results show no correlation, i.e. high Ba concentrations are invariably associated with mean Sr concentrations. This suggests that the observed effect is a result of dietary rather than metabolic factors.

Observed Ratios. The skewed distribution makes it somewhat difficult to assess the results. It is at present not certain whether an overall average should be disregarded. The decision might be easier if Sr^{90} results were known to correlate with high natural Sr.

Two values for the O.R. (bone/diet) are consequently obtainable for each age group. These are:

Sr/Ca O.R.'s (bone/diet)

Age Group	No. of Samples	Mode	Mean
0-20 years	25	0.24	0.24
20-70 "	56	0.22	0.26
0- 5 "	100 Teeth	-	0.26

As you can see, all values for the observed ratio are in the neighborhood of 0.25 with the mean about 15% higher than the mode.

Similar calculations for Ba give:

Ba/Ca O.R.'s (bone/diet)

Age Group	Mode	Mean
0-70 year	0.05	0.07

The spread between the mode and the mean Ba O.R.'s is about twice as large as for Sr.

If we now compare Ba with Sr:

Ba/Sr O.R.'s (bone/diet)

Age Group	Mode	Mean
0-70 year	0.25	0.28

You will note that the Ba/Sr and Sr/Ca O.R.'s are remarkably similar. It is interesting to speculate that this trend should be carried over the last member of the series, Ra, which might be expected to have an O.R. (Ra/Ca) approximately 1/4 of that reported for the Ba/Ca ratio.

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THE EFFECT OF DEPOSITION RATE AND CUMULATIVE SOIL LEVEL ON THE CONCENTRATION OF STRONTIUM-90 IN U. S. MILK AND FOOD SUPPLIES*

Harold A. Knapp
Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission
Washington, D. C.

The object of this report has been to seek an approximate, quantitative description of the way in which the Sr^{90} level of the average United States diet has been dependent on the average cumulative level of Sr^{90} in the soil, and upon the average Sr^{90} deposition rate. Such other observations from the data as appear justified have also been noted. For quantitative treatment, the milk levels are expressed in $\mu\mu c$ Sr^{90}/l, food levels in $\mu\mu c$ Sr^{90}/g Ca (or strontium units), soil levels as mc $Sr^{90}/mi.^2$, and deposition rates as mc $Sr^{90}/mi.^2/mo$.

Diet levels of Sr^{90} are interesting primarily as a means of estimating current and future bone levels. A major uncertainty in predicting future diet levels from nuclear testing conducted through 1958 has been that of estimating in what way past diet levels have been influenced by the rate of Sr^{90} fallout, and in what way by the accumulated amounts of Sr^{90} in soil.

Milk is the component of the U. S. diet which has been most comprehensively measured for Sr^{90} to date. Even though it has been estimated as contributing less than half of the total amount of this nuclide in the average adult diet, it appears to reflect the average Sr^{90} content of this diet. For very young infants it might be taken to represent the entire diet. A study by Consumers Union (1) of whole diets in 8 U. S. cities in November 1959 found that the average strontium unit level of the diets was 1.2 times that of the average milk. The data for the Consumers Union study are shown in Table 1. A three

* During the conference only a very brief summary was presented. The paper included here is reproduced in edited form without appendices from Division of Biology and Medicine, U. S.Atomic Energy Commission report TID-13945, October 1, 1961. 53pp.

city diet by the U. S. Atomic Energy Commission's Health and Safety Laboratory (HASL) (2) in the spring and fall of 1960 gave an average level for the diet which was 1.5 times that of the average milk. The data from the HASL study are shown in Table 2.

It is far from certain that future average Sr^{90} levels in whole diet and milk will continue to have a ratio in the range of 1.2 - 1.5, although lacking any additional data, such an assumption is perhaps not unreasonable. If then it is possible to establish from past data an approximate quantitative relation between average Sr^{90} soil levels and deposition rates (as independent variables), and milk levels (as the dependent variables), there is some basis for using projected soil levels and rate data to estimate future levels of Sr^{90} in milk and the whole diet.

TABLE 1

Sr^{90} in Milk and in Whole Diets*

City where Sample taken	Strontium units in milk	Strontium units in total diet	Diet/ Milk
Washington, D. C.	9.3	11.2	1.20
Atlanta, Georgia	8.0	11.5	1.44
Louisville, Kentucky	10.0	9.4	.94
New Orleans, Louisiana	16.2	16.7	1.03
Boston, Massachusetts	9.0	15.5	1.72
St. Louis, Missouri	12.1	15.9	1.31
Brooklyn, New York	10.9	12.2	1.12
Pittsburgh, Pa.	11.6	13.2	1.14
Average	10.9	13.2	1.23

* Samples taken by Consumers Union in November 1959(1)

It has been evident for some time that the relatively heavy Sr^{90} fallout rate which occurred over the United States during March, April, and May of 1959 caused temporary high concentration of Sr^{90} in U. S. milks, although until recently it has not been possible to say with much assurance what fraction

of that increase was in some way directly related to the deposition rate -- presumably resulting in direct foliar contamination -- and what fraction due to the cumulative levels of Sr^{90} in the soil and the root mat system. The major difficulty in untangling these influences has been the lack of quantitative data on Sr^{90} deposition rates and soil levels, and on milk levels from the same area. The most comprehensive milk analysis programs have been those of the U. S. Public Health Service (PHS). However, in none of the 5-10 milkshed areas where milk has been routinely sampled since May 1957 (The USPHS Raw Milk Sampling Program) have there also been measurements of the Sr^{90} deposition rate or measurements of Sr^{90} in the soil*, and only in two of the metropolitan areas seved in part by milk from the sampled milksheds, namely New York and Salt Lake City, have there been both rate and soil measurements. There is a routine 4 month timelag in the reporting of results from the analysis of the USPHS milk samples. The Sr^{90} monthly deposition rates measured by HASL are currently analyzed and reported about 9 months after collection. Many of these samples have been lost, some give large variations in the measured value when the measurements are repeated, and since May 1960 the data have been reported only for bimonthly periods.

In addition to the difficulties due to the available data (or the lack of it), nature has seemingly conspired to make the information which does exist quite difficult to interpret. Different locations have had widely differing Sr^{90} deposition rates, cumulative levels, and amounts of rainfall. Soils vary widely in their calcium content, and presumably therefore in the Sr^{90} uptake by plants for a given cumulative level of Sr^{90} in the soil.

*There have been rate measurements in New York, Chicago, and Salt Lake City, but the milksheds of these areas cover several counties between 50 and 150 miles away from the metropolitan areas.

TABLE 2

Sr^{90} in Milk and Whole Diet*

City	Date	Strontium Units Milk	Strontium Units Whole Diets	Diet/ Milk
New York, New York	March 1960	9.1	11.4	1.25
New York, New York	June 1960	10.1	12.3	1.22
Chicago, Illinois	May 1960	6.2	11.1	1.80
Chicago, Illinois	Sept. 1960	5 1	7.6	1.49
San Francisco, California	March 1960	4.0	5.3	1.32
San Francisco, California	August 1960	1.8	3.1	1.72
Average		6.05	8.5	1.5

* Results from HASL Tri-City Diet Study

Rainfall may be expected to influence the uptake due to cumulative level as well as the deposition rate itself. Distribution of the cumulative level in depth, and its rate of change with time varies from soil to soil, and so on. In view of the complexities of the problem and the incompleteness of the data, it may very well be that a relation of the type sought here is far too simple to give valid projections of future diet levels, or to apply to different fallout situations than the one from which it was derived. On the other hand, lacking other evidence, it is not unreasonable to use the quantitative relation developed here between the average level of Sr^{90} in U. S. raw milk, the average U. S. deposition rate of Sr^{90}, and the average level of Sr^{90} in the soil as a basis for estimating the average levels of Sr^{90} which might be expected in U. S. milk as the result of varying degrees of atmospheric testing. One would not expect the relation developed to provide a meaningful estimate of Sr^{90} milk levels in the U. S. in a period immediately following a nuclear attack if a majority of the dairy cattle had been killed. It might, however, be possible to develop similar relations with different coefficients to apply to various milk producing areas where the cattle had survived a nuclear war and were able to obtain their principal food by grazing.

Previous Models

In their testimony at the 1959 Congressional Hearings on Fallout from Nuclear Weapons Tests, Langham and Anderson (4), describe a method for predicting average milk concentration from average soil levels, i. e. neglecting the effect of fallout rate. Following Libby, this model assumes an average os 20 g of available calcium per $ft.^2$ of soil to a depth of 2.5 in., so that 1 mc $Sr^{90}/mi.^2$ would give 1.8 $\mu\mu c$ Sr^{90}/g Ca in the soil to this depth. They further assume that the discrimination is such that the Sr^{90} to calcium ratio in milk is 0.13 times Sr^{90} to calcium ratio in soil. This gives the relation

$$\mu\mu c\ Sr^{90}/g\ Ca \text{ in milk} = (0.234)\ (mc\ Sr^{90}/mi.^2 \text{ on the ground}).$$

Since 1 liter of milk has approximately 1.1 g of Ca, one may write

$$\mu\mu c\ Sr^{90}/1 \text{ milk} = (0.258)\ (mc\ Sr^{90}/mi.^2)$$

$$\simeq 1/4\ (mc\ Sr^{90}/mi.^2).$$

In an article in 1960, Burton, Milbourn, and Russell (5) conclude with respect to milk in the United Kingdom that

> "At the present time (1958) about 20 per cent of the strontium 90 in milk is due to absorption from the soil; furthermore, it appears that if the present cumulative deposit were more thoroughly incorporated in the soil, it would lead to a level in milk of approximately 12 per cent of that now due to world-wide fallout."

Burton, et al. did not suggest that this conclusion applied to any situation other than that of the United Kingdom in 1958. Their conclusion was generalized by Kulp, Schulert, and Hodges (7):

> "Evidence has been accumulating to show that direct uptake of strontium-90 from rain by plants is the dominant mechanism by which strontium-90 enters the diet of human beings. The most definitive experiment was done by R. S. Russell's group at the Agricultural Research Council Radiobiological Laboratory in the United Kingdom (6). In this it was shown from the activity in milk produced in England and Wales in 1958, that no more than 20 percent of the strontium-90 in the 1958 diet could be accounted for by uptake by grass through the soil, and thus that 80 per cent must have been taken into the grass directly from rain. Thus, the rate of fallout under weather conditions of 1954 through 1959 was much more important than the cumulative deposit in determining the dietary level. Since the rate of fallout has already dropped below the 1958 level, it appears that the peak of strontium-90 concentration in the world diet has already occurred, in 1959, and the peak in newly depositing bone will occur this year. (Emphasis added).
>
> "From the inventory values, and the residence time indicated above, and the value of 20 per cent for the contribution of the cumulative deposit to the level of strontium-90 in the skeletons of people of any age as a result of nuclear test to date."

The conclusions reached in this paper suggest that it was an error for Kulp, Schulert, and Hodges to have accepted the conclusions of Burton, Milbourn, and Russell with respect to the Sr^{90} content of milk products in England and Wales in 1958 and applied them without qualification to total diets throughout the entire world from 1954 thru 1959.

The Data

I. The Milk Data. The milk analyzed under the PHS Raw Milk Sampling Program is collected once each month by a local public health representative from a milk receiving station which supplies milk to a metropolitan area and whose source of supply is a milkshed area the size of several counties. Seven of

9 metropolitan areas by which the sample is identified in the monthly reports of the PHS are not located in the milkshed from which the milk was obtained, and may in fact be a considerable distance from the milkshed. The samples from New York and St. Louis, Missouri, for example, come from counties approximately 150 mi. away from these cities. A more detailed description of the PHS milk sampling program is given in Appendix I of TID-13945, and in (3).

The results of the monthly milk analysis (3) for each of the milkshed areas are plotted in Figs. 1a, 1b, and 1c, and the average for all areas is plotted in Fig. 2. The basic data are reproduced as Appendix II, of TID-13945. It should be noted that each month's milk sample was not always collected on the same day in each of the metropolitan areas, and that each metropolitan area's monthly milk sample was not always collected on the same day of the month for every month. This fact was not taken into account when Figs. 1 and 2 were plotted; the monthly values shown there are all treated as if they were collected on the middle of each month. Whence estimates of, say, the time at which the maximum reported milk values occurred may be off by half a month in either direction.

Use of the PHS milkshed data to draw conclusions as to the average level of Sr^{90} in U. S. milk supplies since mid-1957 raises the question as to how well the PHS data represent the U. S. average, particularly in the first year when the only information came from the New York, Cincinnati, St. Louis, Salt Lake City, and Sacramento milksheds. The information available suggests that, lacking more detailed data, it is not an unreasonable estimate. St. George, Utah, and Overton, Nevada, included in the PHS program because of their proximity to the Nevada Test Site, have <u>not</u> been included in the milk data considered here on the grounds that they would weight the limited data too heavily toward an area of the country too dry to be representative. (These milksheds have had a much lower than average concentration of Sr^{90} in milk.) If, in Fig. 3, one continues to plot the average milk levels of the original 5 stations after the addition of the Atlanta, Austin, Chicago, Spokane, and Fargo/Moorehead milksheds, the agreement is quite close. The newly established PHS program for sampling processed milk at 60 points of public distribution throughout the U. S. has been reporting data (3) since July 1960. The average of these results is also shown in Fig. 2, and is seen to be in no substantial disagreement with the average of the milkshed data.

It should be noted that some of the first samples reported here for the Salt Lake City milkshed are not in agreement with those previously published by the PHS. The data plotted here include the corrected values provided by the PHS.

II. The Sr^{90} Deposition Rate Data. The monthly deposition of Sr^{90} is measured by HASL at 29 locations throughout the 48 continental States of the United States. The location of the measuring stations, the two principal methods for measuring Sr^{90} deposition, and discussions of the accuracy of measurement are given in the quarterly summary reports (2).

FIGURE 1A

PUBLIC HEALTH SERVICE MILKSHED DATA

FROM RAW MILK SAMPLING PROGRAM

STRONTIUM -90 CONCENTRATIONS IN μμc / LITER

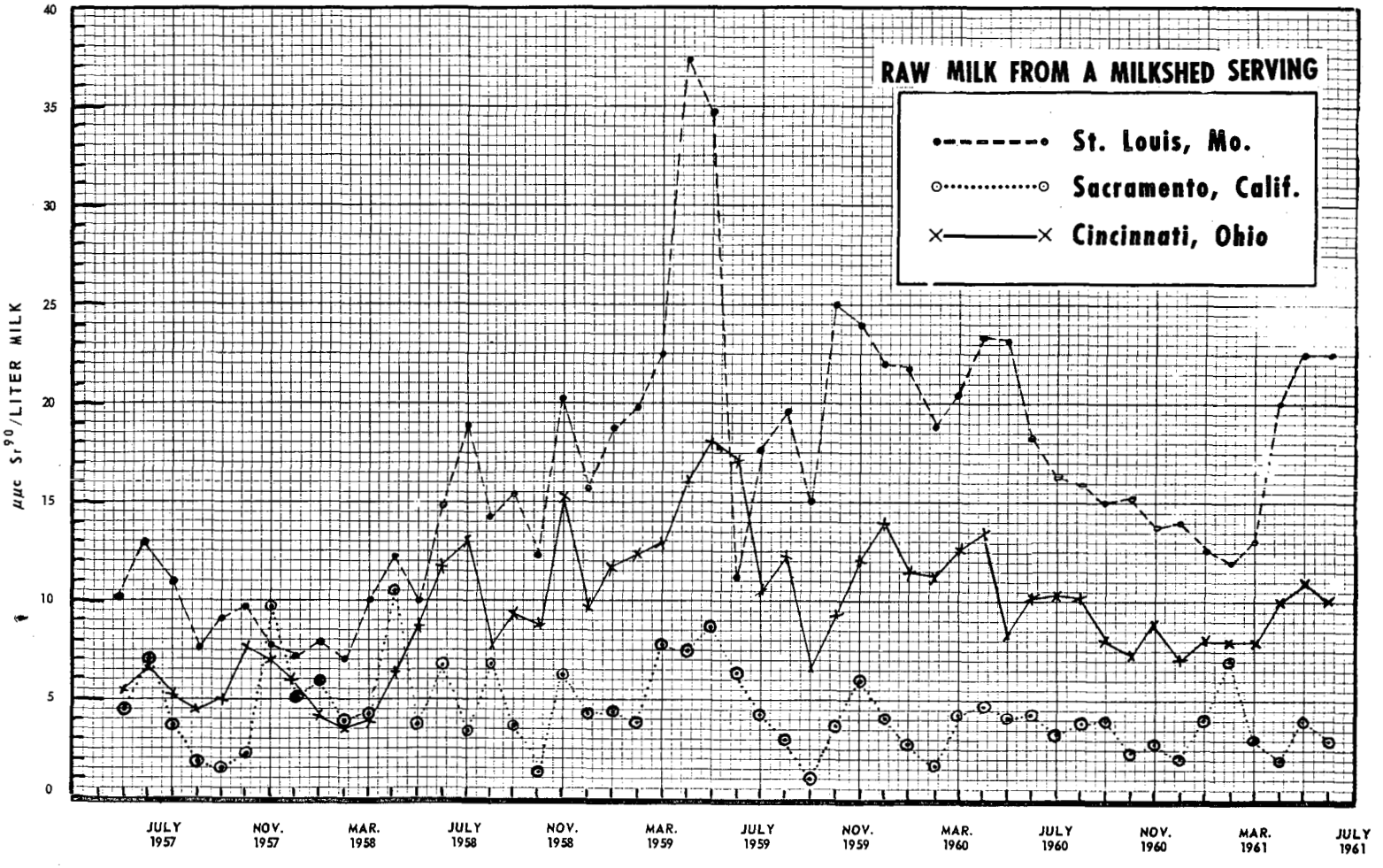


FIGURE 1B

PUBLIC HEALTH SERVICE MILKSHED DATA

FROM RAW MILK SAMPLING PROGRAM

STRONTIUM - 90 CONCENTRATIONS IN μμc / LITER

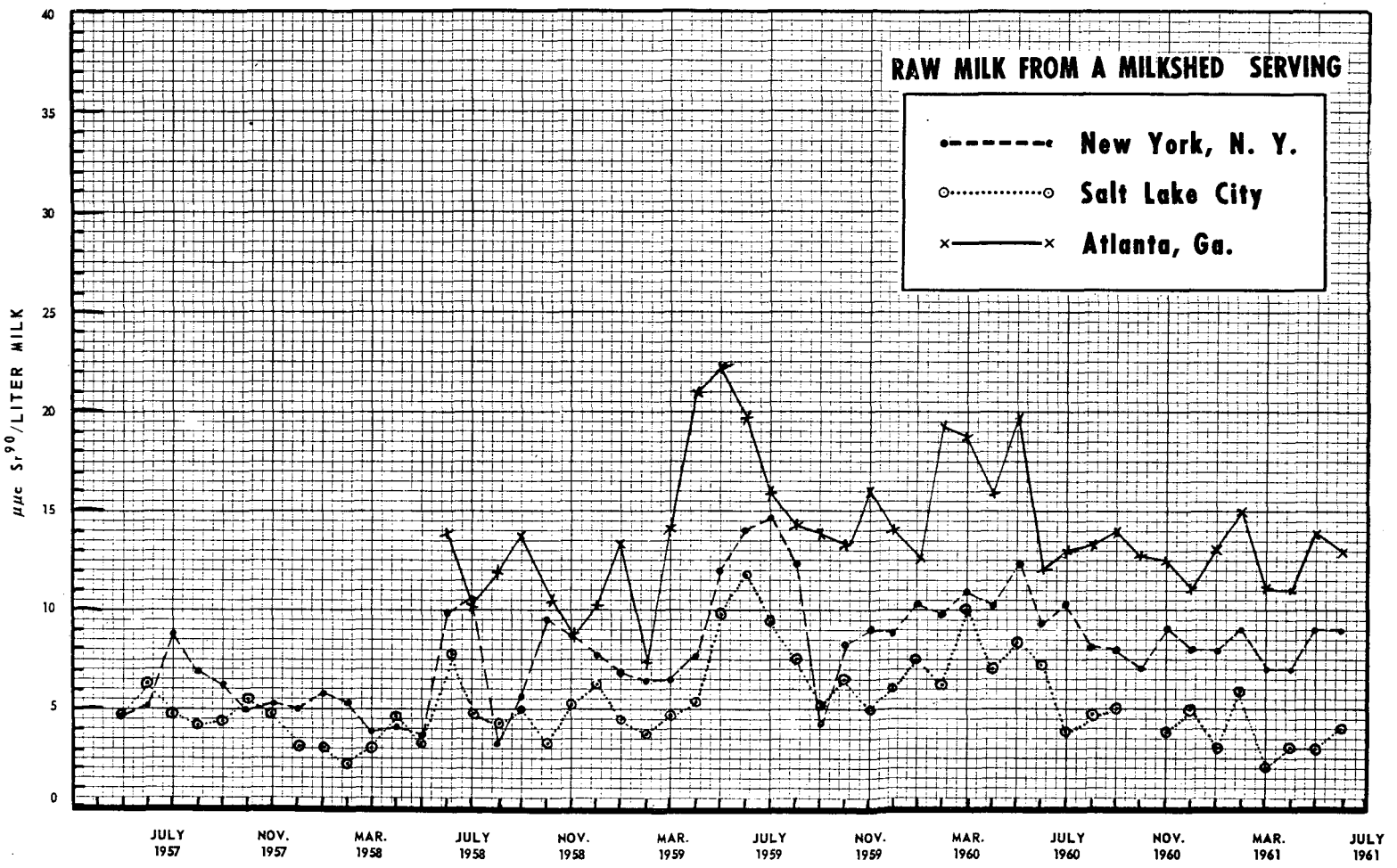


FIGURE 1C

PUBLIC HEALTH SERVICE MILKSHED DATA

FROM RAW MILK SAMPLING PROGRAM

STRONTIUM - 90 CONCENTRATIONS IN μμc / LITER

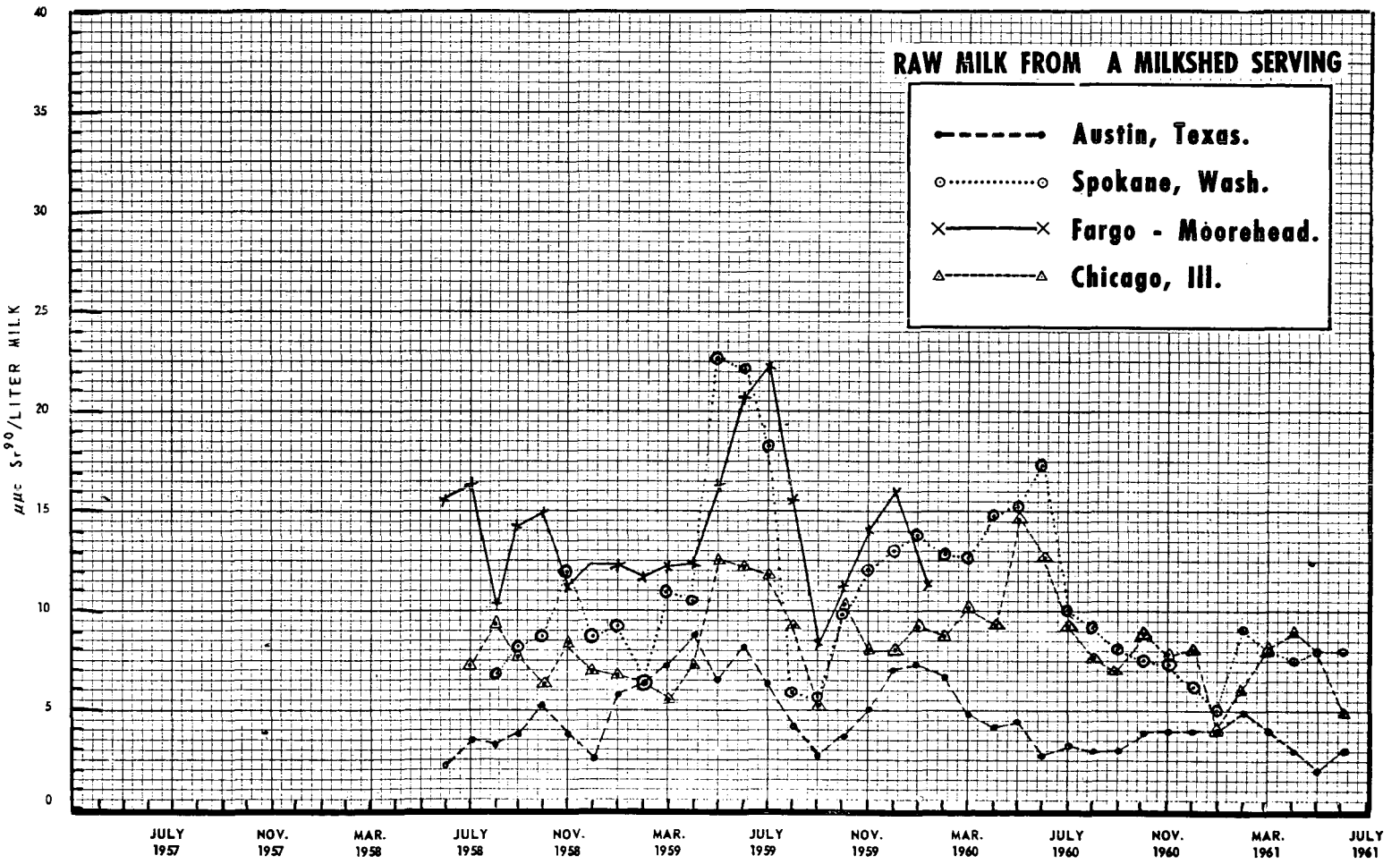
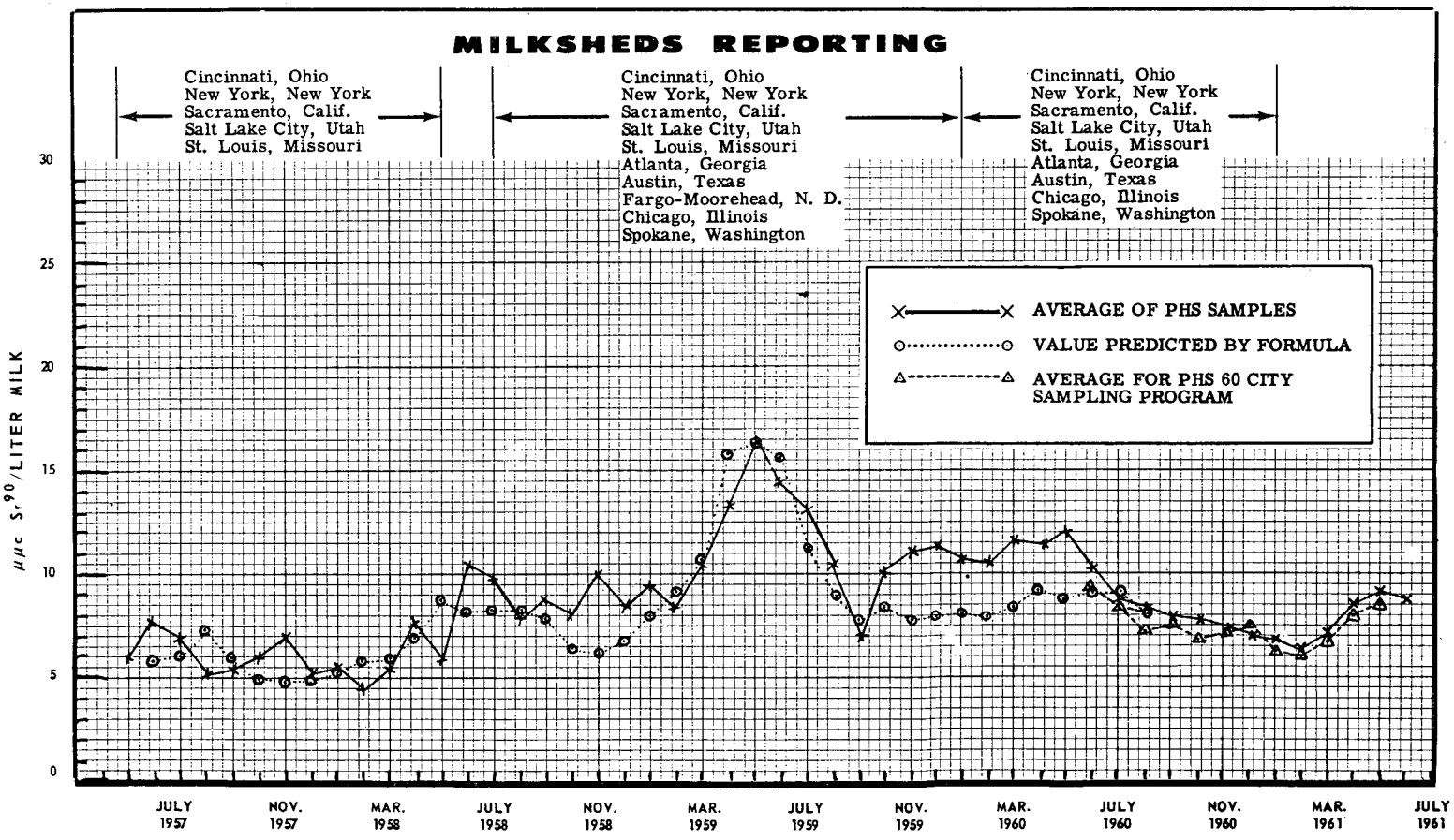


FIGURE 2

AVERAGE MONTHLY CONCENTRATIONS OF SR 90 IN MILK SAMPLED IN PUBLIC HEALTH SERVICE MILKSHED AREAS THROUGHOUT THE U. S., TOGETHER WITH VALUE COMPUTED FROM THE FORMULA

$$\left[\begin{array}{l}\text{average strontium-90 level in U. S. milk supplies, expressed in } \mu\mu c/\text{liter}\end{array}\right] = \frac{1}{9}\left[\begin{array}{l}\text{average cumulation strontium-90 level in U. S. soil, expressed in mc/mi}^2\end{array}\right] + 2.6\left[\begin{array}{l}\text{average strontium-90 deposition for the preceding month, expressed in mc/mi2/mo}\end{array}\right]$$



For the purpose of this analysis, the average monthly Sr^{90} deposition on the U. S. (in mc $Sr^{90}/mi.^2$) was computed using available data from 10 fallout measuring stations located as close as possible to each of the metropolitan areas for which the PHS milkshed data are reported. The PHS metropolitan areas and the HASL collection stations selected are:

PHS Metropolitan Areas	HASL Collection Station
1. Atlanta, Georgia	Birmingham, Alabama
2. Austin, Texas	Houston, Texas
3. Chicago, Illinois	Chicago, (Lemont) Illinois
4. Cincinnati, Ohio	Pittsburgh, Pennsylvania
5. New York, New York	New York, New York
6. Sacramento, California	West Los Angeles, California
7. Salt Lake City, Utah	Salt Lake City, Utah
8. Spokane, Washington	Medford, Oregon
9. St. Louis, Missouri	Tulsa, Oklahoma
10. Fargo, North Dakota	Vermillion, South Dakota

These two sets of cities are shown in Fig. 3. It is seen that only New York, Chicago, and Salt Lake are available for both sets of data, although in none of these three cases is the pot collection station located within the milkshed area, and in the case of New York City, it is separated by 150 mi. The Sr^{90} monthly deposition data from the 10 cities selected is given in Appendix III of AEC report TID-13945. One difficulty with these data is the frequent loss samples, particularly from the Lemont (Chicago) collection station.

The computed monthly average deposition rate is plotted in Fig. 4. The monthly rates from New York, Chicago, and Salt Lake City are shown in Fig. 5.

III. The Sr^{90} Cumulative Soil Level Data. The latest reported comprehensive measurements of Sr^{90} in U. S. soils were made in 1959 by U. S. Department of Agriculture (USDA), and are reported by Alexander, et al. (6). From these data, it was possible to select one or two stations located at or near one of the PHS milkshed metropolitan areas, and/or at or near one of the HASL fallout collection stations previously selected as near the milkshed metropolitan area. The soil sampling stations selected are shown in Fig. 7. The Sr^{90} content of the soils measured at these points is shown in Appendix IV of TID-13945. The average of the 14 stations so

HASL STATIONS FOR MEASURING THE MONTHLY DEPOSITION OF STRONTIUM-90 SELECTED FOR CALCULATION OF THE AVERAGE MONTHLY DEPOSITION RATE ON THE U. S., AND METROPOLITAN AREAS RECEIVING MILK FROM MILKSHED AREAS UTILIZED IN

FIGURE 3

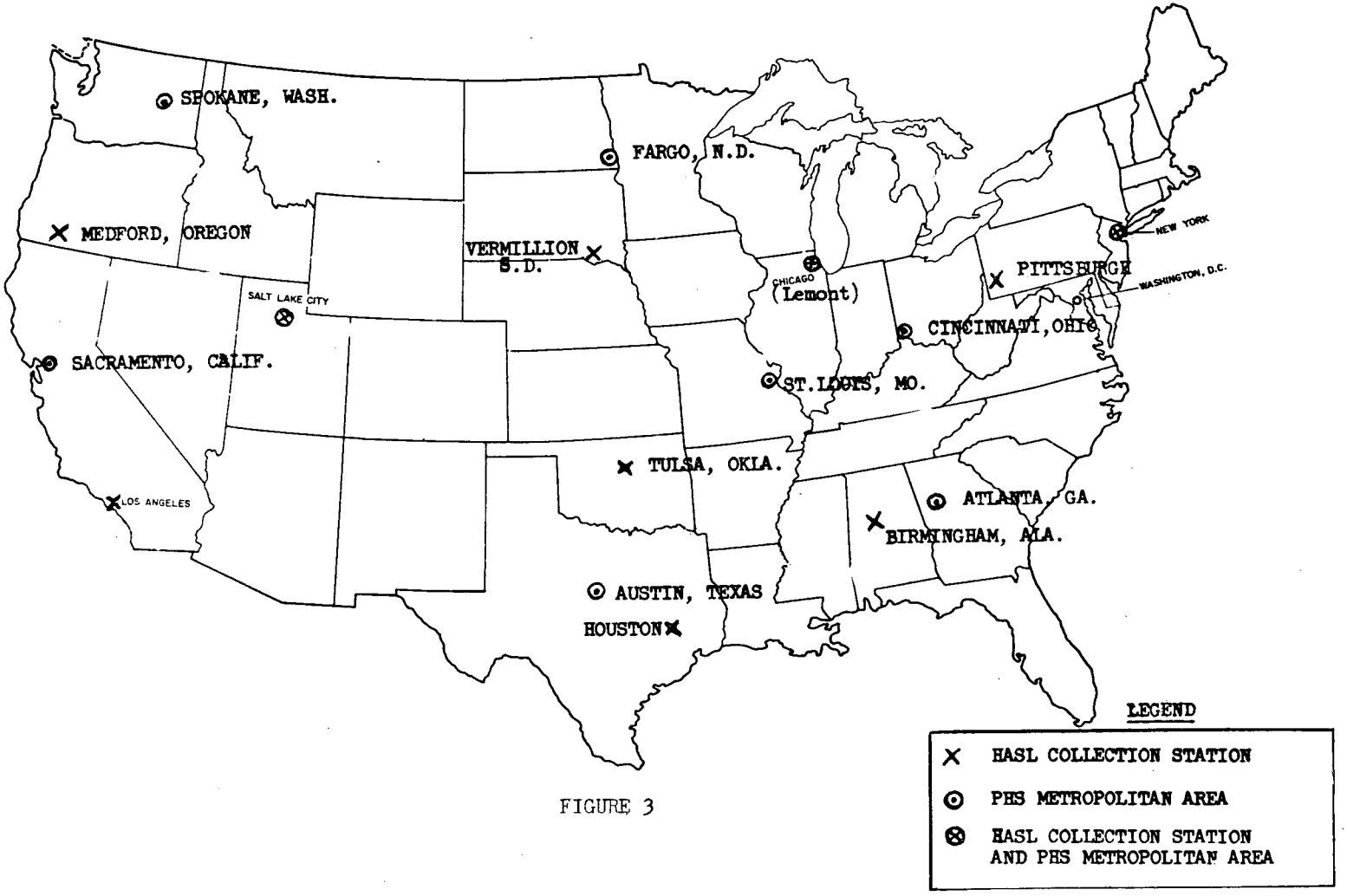


FIGURE 3

STRONTIUM 90

FIGURE 4

DEPOSITION RATE ON THE UNITED STATES

AS DETERMINED FROM THE AVERAGE OF THE MONTHLY DEPOSITION MEASUREMENTS MADE AT THE 10 HASL COLLECTION STATIONS SHOWN IN FIGURE 3.

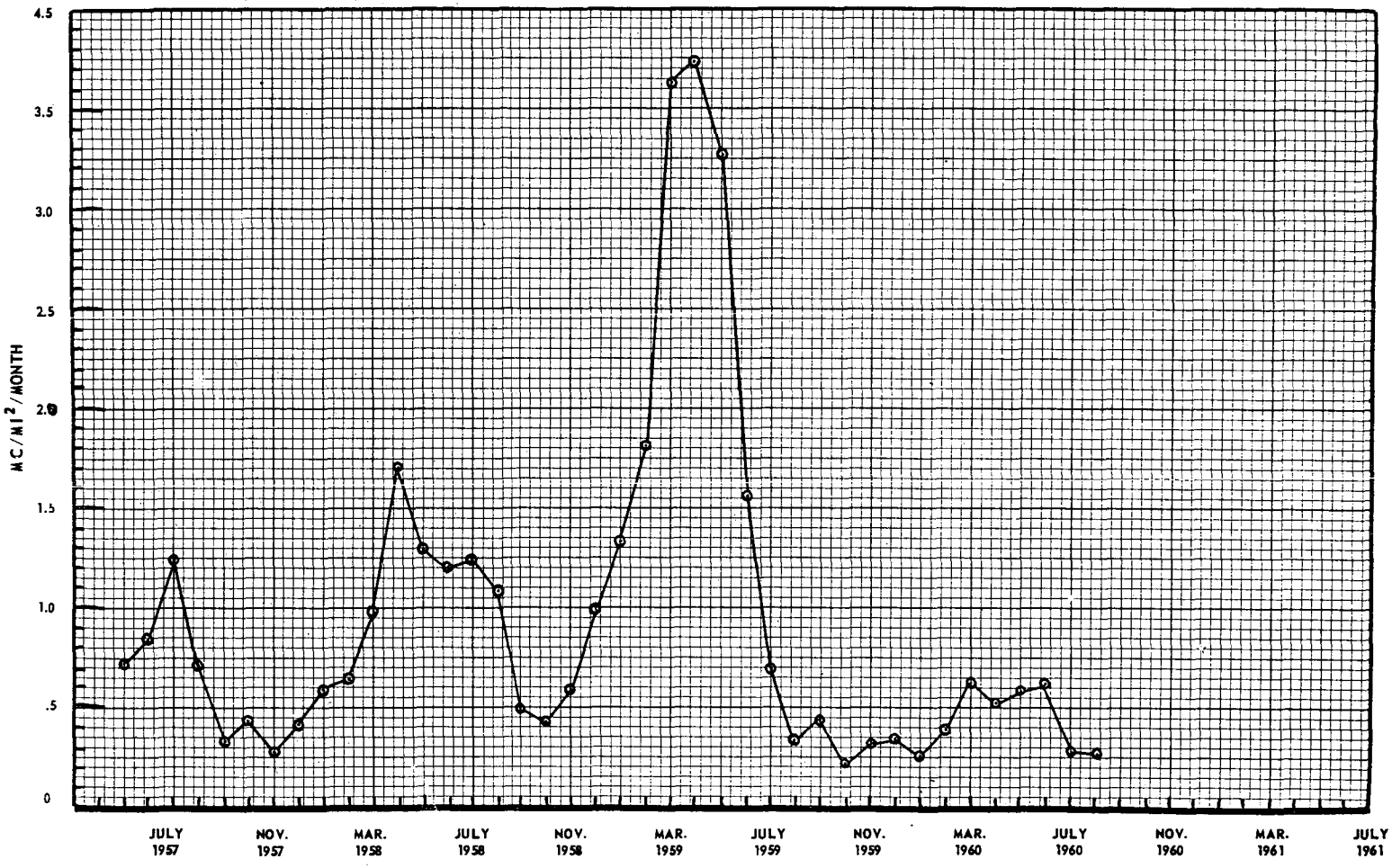
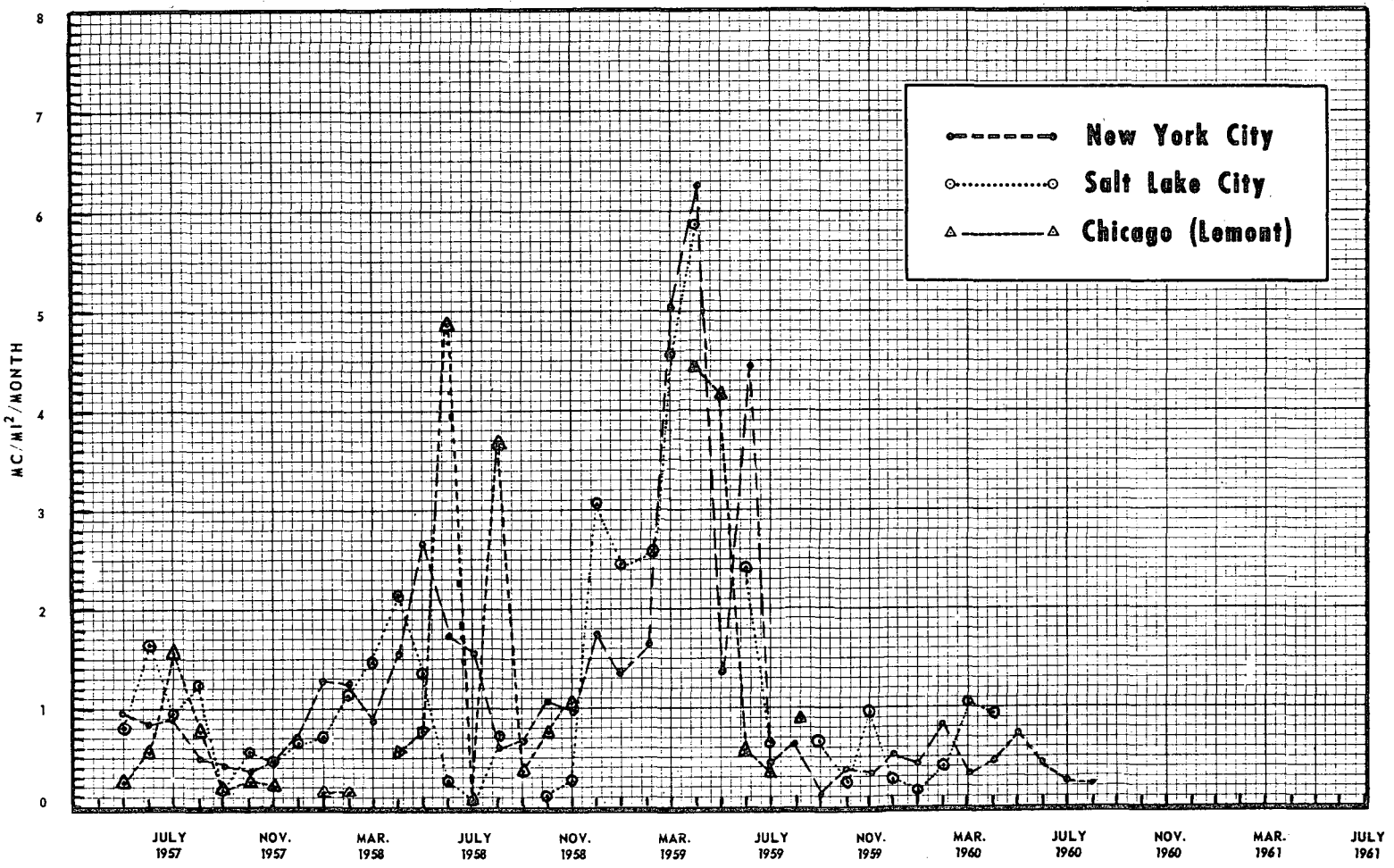


FIGURE 5

HASL MEASUREMENTS
OF THE STRONTIUM - 90 MONTHLY DEPOSITION RATE
FOR NEW YORK CITY, SALT LAKE CITY AND CHICAGO (LEMONT)



chosen is taken to represent the average cumulative Sr^{90} level in the United States in the fall of 1959. From these measurements it is estimated that the average Sr^{90} content of U. S. soils was 65 mc $Sr^{90}/mi.^2$ as of November 1957. The Sr^{90} monthly deposition rates shown in Fig. 4 (and Appendix III of TID-13945) were then integrated to give the cumulative deposition since May 1959, and the resulting curve was superimposed on the soil measurements in order to pass thru a valve of 65 $mc/mi.^2$ in November 1959. The results are plotted on Fig. 6.

Analysis and Conclusions

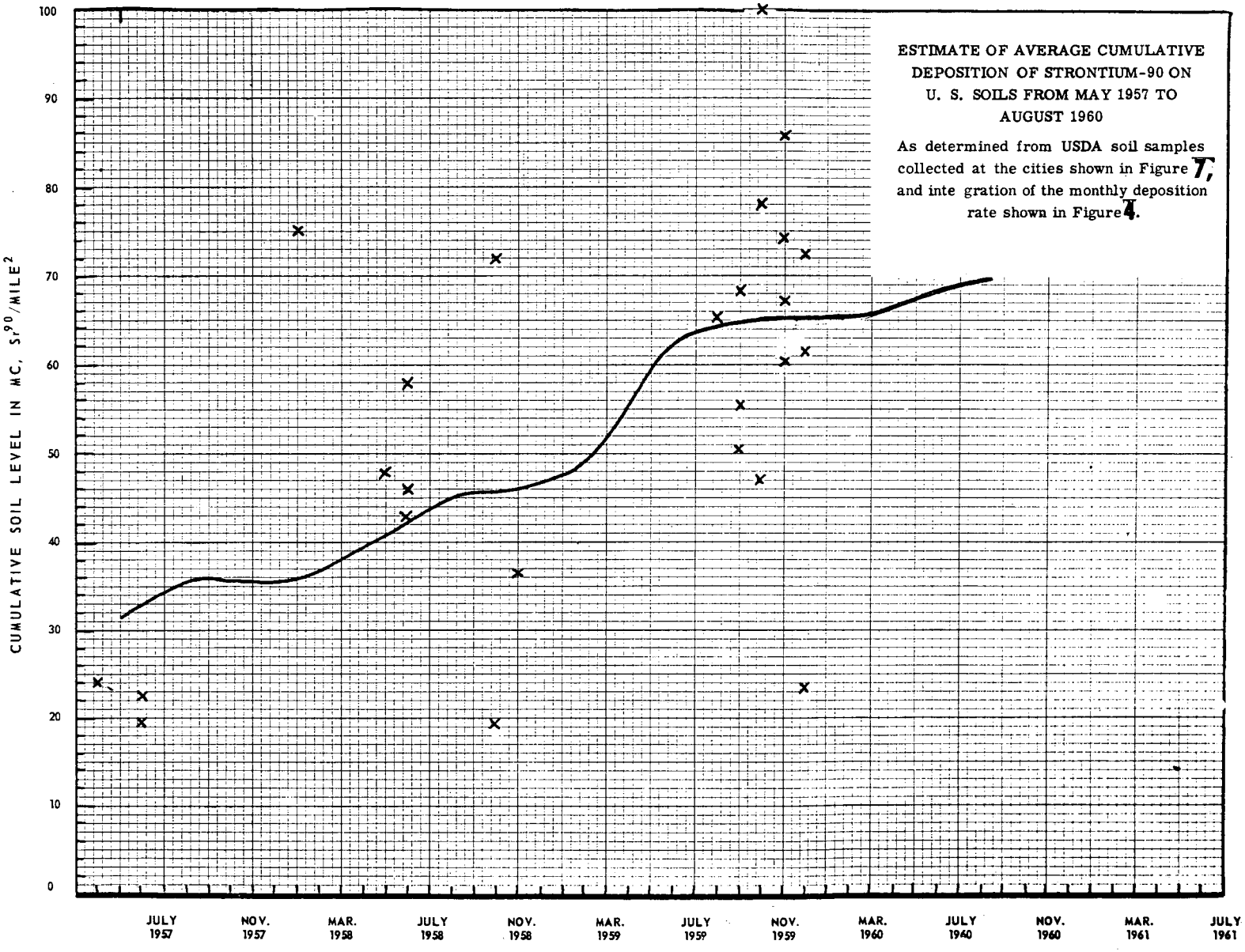
1. For a 8 month period starting in June 1960, during which time the U. S. Public Health Service has measured the Sr^{90} content of raw milk in milksheds serving Atlanta, Georgia; Austin, Texas; Chicago, Illinois; Cincinnati, Ohio; New York, New York; Sacramento, California; Salt Lake City, Utah; Spokane, Washington; and St. Louis, MIssouri, and of processed milk at 60 distribution points within the United States, the average monthly Sr^{90} levels of the two sets of data agree within 20%. This suggests that the average Sr^{90} content of milk from the selected PHS milksheds can be used as an approximate indication of the average Sr^{90} content of the milk consumed in the continental U. S. before the 60 station network was established.

2. Since July of 1958, the Sr^{90} concentration in the milk samples taken each month in the PHS milksheds listed above has varied from station to station by a factor of about 7. The highest concentrations have been almost observed in the St. Louis, Missouri, milkshed (December 1960 = 14 $\mu\mu c/l$), and the lowest in Sacramento, California milkshed (December 1960 = 2 $\mu\mu c/l$), with the Cincinnati, Ohio milkshed approximating the average level (December 1960 = 7 $\mu\mu c/l$). The highest monthly level reported was 37 $\mu\mu c/l$, from the St. Louis milkshed in April 1959. Measurements by the PHS since June 1960, of the Sr^{90} content of processed milk at milk distribution points in 60 cities around the U. S. show that since that time other cities have had levels as high as those reported for the St. Louis milkshed. Presumably these cities also experienced levels as high as those observed in the St. Louis milkshed during the spring of 1959. A comparison of the milk from the St. Louis milkshed with that from the 60 city sampling network is given in Table 3.

3. The Sr^{90} content of the milk from PHS milksheds (Figs. 1 and 2) and the Sr^{90} monthly deposition rates reported by HASL (Figs. 4, 5) indicates that, for the most part, both the average milk levels and the individual station milk levels have reached a peak 1 or 2 months following a peak in the average and individyal station Sr^{90} monthly deposition levels.

4. Although Sr^{90} diet levels in individual cities have been found to be up to two times the Sr^{90} milk levels in these cities (on a strontium

FIGURE 6


ESTIMATE OF AVERAGE CUMULATIVE DEPOSITION OF STRONTIUM-90 ON U. S. SOILS FROM MAY 1957 TO AUGUST 1960
As determined from USDA soil samples collected at the cities shown in Figure 7, and inte gration of the monthly deposition rate shown in Figure 4.
CUMULATIVE SOIL LEVEL IN MC, Sr^{90}/MILE^2
100
90
80
70
60
50
40
30
20
10
0
JULY 1957
NOV. 1957
MAR. 1958
JULY 1958
NOV. 1958
MAR. 1959
JULY 1959
NOV. 1959
MAR. 1960
JULY 1940
NOV. 1960
MAR. 1961
JULY 1961

USDA SAMPLING LOCATIONS SELECTED FOR COMPUTATION OF THE AVERAGE CUMULATIVE DEPOSITION OF STRONTIUM-90 ON U. S. SOILS

FIGURE 7

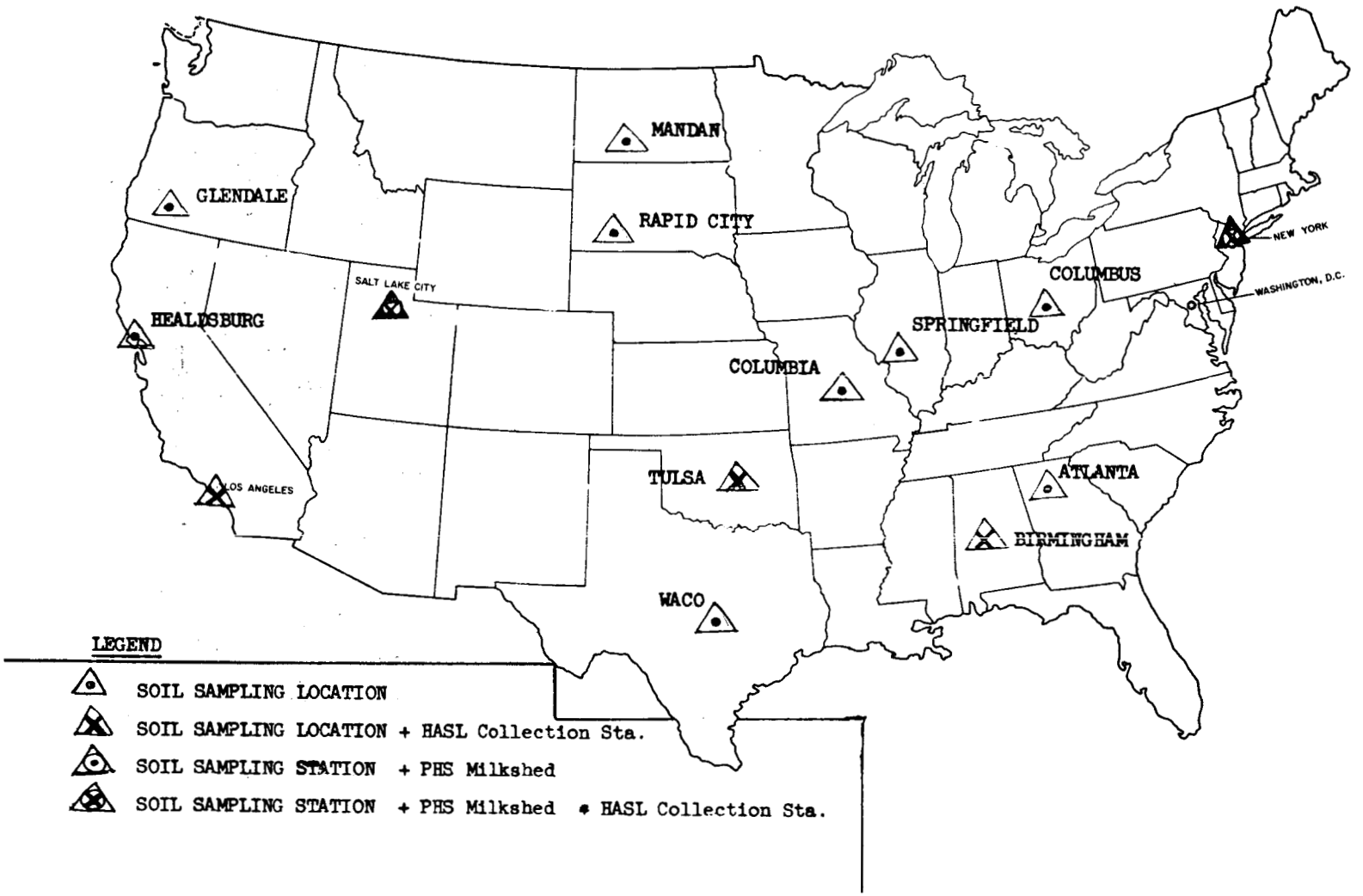


TABLE 3

Comparison of the Sr^{90} Content of Milk from the St. Louis Milkshed with that of Processed Milk Sampled at Distribution Points Within Selected U. S. Cities*

Month Sampled	St. Louis Milkshed μμc Sr^{90}/l	Milk Sampled at Metropolitan Areas μμc Sr^{90}/l	
June 1960	18.1	Boston, Mass.	16
		Charlotte, N. C.	14
		Manchester, N. H.	14
		Pittsburgh, Pa.	16
		Providence, R. I.	18
		Portland, Maine	15
July 1960	16.3	Boston, Mass.	17
		Charlotte, N. C.	16
		Manchester, N. H.	14
		New Orleans, La.	18
		Pittsburgh, Pa.	16
		Portland, Maine	16
		Providence, R. I.	14
August 1960	16.0	Jackson, Miss.	14
		New Orleans, La.	14
		Altanta, Georgia	11
		Providence, R. I.	11
September 1960	15.0	Charlotte, N. C.	13
		Little Rock, Ark.	16
		Memphis, Tenn.	14
		New Orleans, La.	16
October 1960	15.1	Charlotte, N. C.	12
		Charleston, W. Va.	12
		Little Rock, Ark.	12
		New Orleans, La.	12
November 1960	13.7	Boston, Mass.	11
		Little Rock, Ark.	14
		Washington, D. C.	11
December 1960	14.0	Charleston, S. C.	12
		Charlotte, N. C.	12
		Jackson, Miss.	14
		Pittsburgh, Pa.	12
January 1961	13.0	Charlotte, N. C.	10
		Chattanooga, Tenn.	10
		Jackson,Miss.	11
		Little Rock, Ark.	12
		New Orleans, La.	10
		Portland, Maine	10
		Spokane, Wash.	10

*PHS data (3).

unit basis), the average adult diet level appears to be about 1.2 to 1.5 times the average diet level.*

5. It is possible to derive several simple formulae which approximate the relation between (1) the average of the monthly measured levels of Sr^{90} in the PHS milkshed areas (Fig. 2), the average monthly deposition of Sr^{90} on the United States as determined from selected HASL fallout collection stations (Fig. 3), and (3) the average cumulative level of Sr^{90} on the United States as measured at selected USDA soil sampling sites (Fig. 3). Several such formulae were tried and compared with the measured value. One of the simplest relations, and the one which, by eye, gave the best fit to the milk data, was based on the assumption that for any given month the milk level may be considered as the sum of two factors, one proportional to the average Sr^{90} content of the soil (expressed in $mc/mi.^2$ cumulative deposition), and the other proportional to the Sr^{90} deposited during the previous month.

average Sr^{90} level in U. S. milk supplies, expressed in $\mu\mu c/l$	= A	average cumulative Sr^{90} level in U. S. soil, expressed in $mc/mi.^2$	+ B	average Sr^{90} deposition for the proceeding mo., expressed in $mc/mi.^2/mo.$

For such a model, one is free to determine values for the coefficients A and B by inserting the observed average values of the quantitities in the parenthesis for any two months, and solving the resulting simultaneous equations. The calculated values of A and B will, of course, depend upon which two months are selected. Two months which provide great contrast in the Sr^{90} deposition data are the month with the lowest rate (also the latest data) and the month with the highest rate. On this basis the choice made here has been to use the deposition rate data for July 1960 (and hence the corresponding milk and soil data for August 1960) since it is the latest rate information available, and to select as the basis for a second equation, the rate and soil data corresponding to the highest observed average milk level, which occurred in May 1959. From these two sets of data one may thus write

$$8.5\ \mu\mu c/l = A(69\ mc/mi.^2) + B(0.28\ mc/mi.^2/mo.)$$

$$16.5\ \mu\mu c/l = A(61\ mc/mi.^2) + B(3.7\ mc/mi.^2/mo.).$$

Solving these two simultaneous equations one finds

$$A = \frac{1}{9} \ \mu\mu c/l/mc/mi.^2$$

$$B = 2.6 \ \mu\mu c/l/mc/mi.^2/mo.,$$ so that the formulae thus determined becomes

$$\begin{pmatrix}\text{average } Sr^{90} \text{ level in U. S. milk supplies, expressed in } \mu\mu c/l\end{pmatrix} = \frac{1}{9}\begin{pmatrix}\text{average cumulative } Sr^{90} \text{ level in U. S. soil, expressed in } mc/mi.^2\end{pmatrix} + 2.6\begin{pmatrix}\text{average } Sr^{90} \text{ deposition for the proceeding mo., expressed in } mc/mi.^2/mo.\end{pmatrix}.$$

A plot of this expression based on the average cumulative level and deposition rate shown in Figs. 4 and 6, is plotted on Fig. 2. The results are seen to agree reasonably well except for the periods October 1957 through January 1958, and October 1959 through May 1960, during which times the formula predicts levels which are about 30% too low. It is possible that this low value is largely a reflection of the fact that during the winter months most cows outside the south were fed to some significant extent on hay and grains grown during the spring and early summer months of 1958 and 1959, when the rates of fallout and the milk levels both reached peak values.

6. The formula given in 5. above predicts Sr^{90} levels in milk which, with the latest reported Sr^{90} deposition rate of 1/2 mc/mi.^{2}/month or less, are about half as great as those predicted by Langham and Anderson (4) on the basis of the relations

$$\begin{pmatrix}\text{average } Sr^{90} \text{ level in U. S. milk supplies, expressed in } \mu\mu c/l\end{pmatrix} \simeq \frac{1}{4}\begin{pmatrix}\text{average cumulative } Sr^{90} \text{ level in U. S. soil, expressed in } mc/mi.^2.\end{pmatrix}$$

It also predicts a percentage contribution from the cumulative level on the soil significantly higher than the one given by Burton, Milbourn, and Russell (5), for the U.K. in 1958, namely that

> "At the present time (1958) about 20 percent of the strontium 90 in milk is due to absorption from the soil; furthermore it appears that if the present cumulative deposit were more thoroughly incorporated in the soil, it would lead to a level in milk of approximately 12 percent of that now due to world wide fallout."

During 1958 the formula given in conclusion 5. above indicates that for U. S. milk, between 51 and 83% of the Sr^{90} level was due to the cumulative level of fallout, with the average value for the year being 69%. The lowest contributions from the cumulative level during any month, according to the formula, is 40%, and this occurred during April 1959, the month of maximum Sr^{90} fallout. Only in April, May, and June, 1959, does the formula predict a contribution from the cumulative level of less than 50%, and by August 1960, the formula indicates 91% of the total to be due to the cumulative level deposited. The data and these calculations cast considerable doubt on the conclusion of Kulp, Schulert and Hodges (7) that

> "..., the rate of fallout under weather conditions of 1954 through 1959 was much more important than the cumulative deposit in determining the dietary level."

7. Assuming that (1) the diet level in strontium units ($\mu\mu c\ Sr^{90}/g\ Ca$) will remain approximately equal to the milk concentration in $\mu\mu c/l$, (2) that the average, future U. S. Sr^{90} fallout rate from nuclear tests conducted through 1958 will always be less than 0.5 $mc/mi.^2/mo.$, and that (3) within a year or two the fallout rate from these tests will fall off to such an extent that the cumulative level of available Sr^{90} in the soil from weapons tests conducted before the moratorium will disappear at a rate determined primarily by radioactive decay (2-1/2%/year), then the furure average Sr^{90} content of the U. S. diet from these past tests may be estimated to be about

$$7e^{-(t/40)} \text{ strontium units,}$$

where t is the time in years starting, say, in January 1962. If there is appreciable downward displacement of the older Sr^{90} in the soil, or fixation of this nuclide in some form so that it is not available to plants, the expression given must be regarded as an upper limit for the Sr^{90} fraction of the diet due to tests conducted before the 3 year cessation of testing beginning in November 1958.

8. Considerable interest has been expressed in Sr^{90} levels in milk and the whole diet which might be experienced in the United States if a 100 MT bomb were detonated under conditions similar to those which gave rise to the relatively high Sr^{90} deposition rate in the spring of 1959. The conclusions of this paper, together with those of Telegadas and Murayama (8), lead to one such estimate.

Telegadas and Murayama note that for the latitude band 30 - 40° N, about 70 - 80% of the sharp peak in Sr^{90} deposition rate occurring in the spring of 1959 derived from the U. S. S. R. October 1958 series. Since this peak reached a maximum average value over the U. S. of 3.7 $mc/mi.^2/mo.$ in April

1959, it may be deduced that about 2.8 mc/mi.^{2}/mo. of this peak value derived from the 12.5 to 15 MT equivalent to fission products injected into the stratosphere by the U. S. S. R. tests in the fall of 1958(9).

From Fig. 5 it is seen that over the period November 1958 to October 1959 the cumulative average Sr^{90} soil level in the U. S. rose from 46 to 65 mc/mi.^{2} The U. S. S. R. 1958 fall series of tests is assumed here to have contributed from 70 to 80% of this increase, or about 14 mc/mi.^{2} The present cumulative level is about 70 mc/mi.^{2}, and the rate for the year preceding the resumption of testing by the U. S. S. R. averaged approximately .5 mc/mi.^{2}/mo.

If a 100 MT weapon deriving 50% of its energy from fission were detonated in October 1961 under such conditions that its fallout were deposited in the same manner as the fallout from the U. S. S. R. 1958 fall series, then assuming that the earlier series released 14 MT of fission products equivalent for global distribution, and that only the atmospheric radioactivity from a 100 MT device were added to that existing before testing resumed in September 1961, one can estimate that a 100 MT bomb would increase the cumulative level of Sr^{90} in the soil during the year from October 1961 to November 1962 by about $\frac{50}{1} \times 14 = 50$ mc/mi.^{2}, and that the fallout rate would reach a peak of about $\frac{50}{1} \times 2.8 = 10$/mc/mi.^{2}/mo. in April 1962. The month-by-month Sr^{90} deposition rate and cumulative level under the conditions postulated would be approximately as shown in Table 4.

The maximum projected Sr^{90} level in milk under the assumed conditions of a 100 MT burst is calculated to occur in May 1962, at a level of 37.5 μμc/l; the average level projected for the one year period November 1961 to October 1962 is 21.8 μμc/l, and the level at the end of this year is 16.3 μμc/l.

Since milk levels in one of the milksheds serving St. Louis, Missouri, and at several distribution centers for processed milk throughout the U. S. have in the past experienced levels roughly twice the U. S. average, one could expect several cities to experience levels of Sr^{90} in milk in the range of 60 to 80 μμc/l for a month or two as the result of a 100 MT bomb, provided the fallout were deposited in a manner similar to that from the U. S. S. R. fall series of tests in 1958.

Lacking any other evidence, it may be assumed that average, adult whole diet levels, expressed in strontium units, would be in the range of 1.1 to 1.4 times the milk levels expressed in μμc Sr^{90}/l, and in the 1.2 to 1.5 times the milk level expressed in μμc Sr^{90}/g Ca.

TABLE 4

Estimate of Sr^{90} deposition level and deposition rate and the resulting average level of Sr^{90} in U. S. milk if a 100 MT device were fired in the atmosphere in October 1960 underfiring conditions similar to those employed in the U. S. S. R. series of the fall of 1958

Date	Calculated Cumulative Level ($mc/mi.^2$)	Calculated Monthly Deposition ($mc/mi.^2$ for previous mo.)	(1/9 X $mc/mi.^2$)	(2.6 X mc/mi^2/mo.)	Projected Milk Level ($\mu\mu c/l$)
November 1961	71.1	1.10	7.9	2.86	10.76
December 1961	72.6	1.49	8.06	3.88	12.04
January 1962	75.3	2.66	8.38	6.92	15.30
February 1962	78.8	3.50	8.76	9.10	17.86
March 1962	83.6	4.82	9.30	12.50	21.80
April 1962	93.3	9.70	10.40	25.20	35.60
May 1962	103.3	10.00	11.50	26.00	37.50
June 1962	112.1	8.80	12.48	22.82	35.30
July 1962	116.2	4.15	12.91	10.89	23.80
August 1962	118.0	1.82	13.10	7.00	20.10
September 1962	118.9	.83	13.20	2.16	15.36
October 1962	120.0	1.15	13.33	2.99	16.32
Average levels for 1 year period	96.9	4.17			21.8

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STRONTIUM-90 IN NEW YORK CITY AND SAN FRANCISCO DIETS RESULTING FROM THE OCTOBER 1961 SOVIET TEST SERIES

Joseph Rivera
Health and Safety Laboratory
U. S. Atomic Energy Commission
New York, New York

The general problem of the prediction of Sr^{90} levels of diets at a given place may be divided into three distinct problems. These are:

1) Finding what foods contribute most Sr^{90} to the average diet.

2) Relating the contamination of these foods to some measurable fallout parameters.

3) Predicting the values of the significant fallout parameters.

For the last two years, the Health and Safety Laboratory has been conducting an experiment that throws some light on the first of these problems (1). Every three months, all of the foods likely to constitute a typical adult diet are purchased in local stores at New York City, Chicago and San Francisco. The foods are prepared as if for consumption, then composited into 19 categories, ashed, and analyzed for Sr^{90} and calcium. Using consumption figures of the U. S. Department of Agriculture 1955 Household Food Survey, it has been found that by far the largest contribution of Sr^{90} to the average annual diet is from milk and dairy products. Of lesser importance are the contributions from bakery products and whole grain cereals and that from fresh vegetables.

Since the contamination of all foods can be traced to the contamination

of plants, the solution of the second problem depends on an understanding of the mechanisms of contamination of plants with Sr^{90}.

The Sr^{90} in plants comes either from the soil or from direct deposition of fallout on the leaves or other above-ground parts of the plant. Factors effecting the uptake from the soil are the amount of Sr^{90} present and its chemical form and distribution within the soil. Other factors of importance are the chemical and physical features of the soil itself, particularly its calcium content. Sr^{90} in plants due to direct deposition depends on the rate of fallout prior to cutting, the size and shape of the leaves, the stage of the development of the plant at the time of contamination and the amount and kind of rainfall to which the plant has been subjected.

Although these are only some of the factors involved, it is reasonable to assume that the two fallout parameters that are of the greatest significance for prediction purposes are the cumulative deposit of Sr^{90} in the soil, and the fallout rate.

A formula suggested by Scott Russell (2) that takes these facts into consideration is:

$$M = a S + b R$$

where 'M' is the Sr^{90}/Ca ratio of the item under consideration, 'S' is the cumulative deposit of Sr^{90} in the soil, 'R' is the fallout rate, and 'a' and 'b' are approximately constant for a given locale.

Various methods that may be used for determining 'a' and 'b' factors for different systems have been summarized by Middleton (3) as follows:

1) Collect data on 'M', 'S', and 'R' for a given region at different times and determine 'a' and 'b' by multiple regression analysis (least squares fit).

2) Determine 'a' directly by measuring the uptake of the system using Sr^{89} in a field experiment. One 'a' is known, the equation can be solved for 'b' given 'M', 'S' and 'R'.

3) Determine 'b' directly by measuring how much Sr^{89} is retained by plants sprayed in a manner simulating fallout conditions. Then calculate 'a'. Another method for estimating 'b' is from Sr^{90}/Sr^{88} (stable strontium) measurements of various parts of the plants (high ratios on the outer parts are ascribed to direct deposition).

4) Assume R = 0 for sometime when the fallout rate was relatively low and solve for 'a' knowing 'M' and 'S'. Substitute this value of 'a' in the equation for sometime when $R \neq 0$ and solve for 'b'.

The formula described above will be used to predict the Sr^{90} levels of

foods in New York City and San Francisco. First, however, the appropriate factors 'a' and 'b' for the important diet components at the two cities must be found.

Applying, with certain modifications, the methods summarized by **Middleton** to determine the 'a' and 'b' factors for milk at New York City and San Francisco, the following results are obtained:

New York City - $a = 0.07$ ($\mu\mu c\ Sr^{90}/g\ Ca$)/$mc\ Sr^{90}/mi.^2$

$b = 4.6$ ($\mu\mu c\ Sr^{90}/g\ Ca$)/$mc\ Sr^{90}/mi.^2$/month

San Francisco - $a = 0.04$ ($\mu\mu c\ Sr^{90}/g\ Ca$)/$mc\ Sr^{90}/mi.^2$

$b = 4.6$ ($\mu\mu c\ Sr^{90}/g\ Ca$)/$mc\ Sr^{90}/mi.^2$/month

The difference in the 'a' factors at the two sites may be due to differences in the calcium content of the soils of the milkshed pastures of New York City and San Francisco.

The situations at times when the cattle were presumably out to pasture were used in deriving these factors. If data for the entire year are used, the factor 'b' is reduced by more than 1/12 of 4.6 and is estimated by Middleton to be 0.23 ($\mu\mu c\ Sr^{90}/g\ Ca$)/$mc\ Sr^{90}/mi.^2$/year. In applying the formula, the delay in the changes of fallout rate being reflected in changing milk levels of Sr^{90} is accounted for by taking the average of the preceding and current month to compute the appropriate 'R' for the prediction of 'M'.

After milk, the most important contributors of Sr^{90} to the diet are bakery products and whole grain cereals. The variation of these items with the cumulative deposit of Sr^{90} and the fallout rate will be taken to be the same as that of wheat flour and whole wheat.

Since the origin of the flour and whole wheat consumed in New York City and San Francisco is probably the same, only one set of factors, 'a' and 'b', need be computed. From the data available, reasonable choices for these factors are:

flour - 'a' = 0.1 ($\mu\mu c\ Sr^{90}/g\ Ca$)/$mc\ Sr^{90}/mi.^2$
'b' = 34 ($\mu\mu c\ Sr^{90}/g\ Ca$)/$mc\ Sr^{90}/mi.^2$ deposited in June

whole wheat - 'a' = 0.1 ($\mu\mu c\ Sr^{90}/g\ Ca$)/$mc\ Sr^{90}/mi.^2$
'b' = 100 ($\mu\mu c\ Sr^{90}/g\ Ca$)/$mc\ Sr^{90}/mi.^2$ deposited in June

In computing 'b' the fallout rate was taken to be that which would occur in June since this is the time when the wheat is most sensitive to direct deposition. The relatively great dependence of the Sr^{90} content of the

whole wheat on direct deposition is reflected in the different 'b' factors for whole wheat and for flour milled from it. Since a good part of the Sr^{90} directly deposited is removed in the milling process, flour is less sensitive to changes in the fallout rate than whole wheat.

In applying the results of predictions for flour and whole wheat Sr^{90} concentrations, to the annual diets at New York City and San Francisco, it must be recalled that bakery products contain locally produced milk as well as flour and hence the Sr^{90} levels in bakery products are expected to differ somewhat between the two cities.

There are considerably less data availableon a continuous basis on the Sr^{90} content of fresh vegetables. Results from the tri-city (New York City, San Francisco, and Chicago) diet surveys are difficult to use for predicting purposes since locally purchased vegetables may not have been locally produced. The best estimate of 'a' and 'b' are probably those based on Middleton's experiments on the retention of sprayed Sr^{89} on various vegetables. (4) His estimates are 'a' = 0.4 ($\mu\mu$c Sr^{90}/g Ca) /mc/mi.^{2} and 'b' = 0.4 ($\mu\mu$c Sr^{90}/g Ca)/mc/mi.^{2}/yr.

In summary, the choices of 'a' and 'b' factors for the prediction of future Sr^{90} levels in diets at New York City and San Francisco that will be used here are:

TABLE 1

Proportionality Factors

		'a'	'b'
milk	New York City	0.07	4.6 (per mo.)
	San Francisco	0.04	4.6 (per mo.)
flour		0.1	34 (per deposit in June)
whole grain products		0.1	100 (per deposit in June)
fresh vegetables		0.4	0.4 (per yr.)

Before we can proceed with the prediction of Sr^{90} levels in diets at New York City and San Francisco, we must solve the third problem proposed at the beginning of this paper, namely; how can we estimate what the fallout situation will be in New York City and San Francisco in 1962 as a result of the recent Soviet test series?

Fallout from the recent Soviet tests can be estimated, in general, by extrapolation of the results of the Soviet test series of October 1958.

Collins has described in this conference (page 271) what the probably fallout rates of Sr^{90} will be like in New York City 1962. A similar analysis can be used to predict the fallout rates in San Francisco. Since the cumulative deposit of Sr^{90} at these cities can be estimated from soil measurements and monthlyfallout collections up to the present time, the cumulative levels to be expected in 1962 can be calculated.

In a paper written prior to the recent test series, Knapp (5) predicted what effect the fallout from the detonation of a nuclear device having a yield of 100 MT would be, on milk levels in the United States as a whole, if the detonation occurred in October 1961. He estimated that the average increase in the level of Sr^{90} in the United States soils resulting from such a detonation would be 50 $mc/mi.^2$ during 1962. The effect of this deposition of Sr^{90} would be to cause the average levels in milk to be about 21 $\mu\mu c$ Sr^{90}/g Ca with peaks of about 35 in April, May and June.

These estimates seem quite reasonable for the country as a whole. The calculations given below for New York City and San Francisco give an indication of the likely fluctuation about the average that might be expected.

From the estimates of Collins, based on a fission yield of 25 MT, we find that from October of 1961 to October 1962 about 45 $mc/mi.^2$ of Sr^{90} are expected to be deposited in New York City as a result of the recent Soviet test series. In the spring months of 1962, the fallout rate may be as high as 13 mc $Sr^{90}/mi.^2/mo$. Using these data and assuming that the cumulative deposit of Sr^{90} in New York City area on October1961 was about 75 $mc/mi.^2$, the average expected Sr^{90} content of milk in New York City in 1962 is calculated to be about 15 $\mu\mu c$ Sr^{90}/g Ca with peaks in May and June of about 50 $\mu\mu c$ Sr^{90}/g Ca.

A similar analysis of the situation in San Francisco indicates that about 20 $mc/mi.^2$ of Sr^{90} due to the Russian tests will be deposited from October 1961 to October 1962. The fallout rate may be as high as 5 or 6 mc $Sr^{90}/mi.^2/mo$. in March of 1962. The contamination of milk from this fallout should result in average milk levels for the year of about 6 $\mu\mu c$ Sr^{90}/g Ca with peaks in March and April of about 20 $\mu\mu c$ Sr^{90}/g Ca.

To calculate the contamination of wheat and flour due to the Soviet tests, we will take Knapp's estimate (adjusted to 25 MT fission) of an average fallout rate of about 25 mc $Sr^{90}/mi.^2/yr.$ and an average cumulative deposit of 90 mc $Sr^{90}/mi.^2$ in the soil. This would result in wheat levels of about 161 $\mu\mu c$ Sr^{90}/g Ca and flour levels of about 73.

Although local conditions of growth are bound to affect the Sr^{90} levels of fresh vegetables reaching the New York City and San Francisco markets, a rough estimate of these levels may be made using Collin's estimate of the cumulative deposit and fallout rates of Sr^{90} that are expected to occur in 1962. The result of about 54 μμc Sr^{90}/g Ca in fresh vegetables grown in 1962 in the New York City area and about 34 μμc Sr^{90}/g Ca for those grown around San Francisco.

The data are too variable to calculate 'a' and 'b' factors for fresh fruits, and meat, fish, and poultry, but a reasonable estimate of the Sr^{90}/Ca ratio in these diet components is about one-third of that in fresh vegetables for fresh fruit and about one-thirtieth that in fresh vegetables for meat, fish, and poultry.

Combining these estimates to get figures for the average strontium to calcium ratio in typical annual adult diets at New York City and San Francisco in 1962, we obtain the results presented in Table 2.

The estimates of the dietary intakes at the two cities for 1962 are probably somewhat on the high side since they take no account of the consumption of foods produced prior to 1962. The rapidity with which Sr^{90}-contaminated crops reach the urban markets is not known with any certainity but it is hoped that measurements of Sr^{89} in the tri-city food samples purchased in 1962 in 1962 will throw some light on this problem.

Despite the many assumptions made in the prediction of the diet levels, it is unlikely that they will prove to be in error by greater than a factor of 2 or 3 if the fission yield estimates for the Soviet test series are fairly accurate.

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TABLE 2

Sr^{90} in New York City and San Francisco Diets in 1962

		μμc Sr^{90}/ g Ca	g Ca/ yr.	μμc/ yr.
Milk	New York City	15	234	3510
	San Francisco	6		(1404)
Bakery Products and Flour		73	37	2701
Whole Grain Products		161	10	1610
Fresh Vegetables	New York City	54	15	810
	San Francisco	34		(510)
Fresh Fruit	New York City	18	15	270
	San Francisco	10		(150)
Meat, Fish & Poultry	New York City	2	55	110
	San Francisco	1		(55)
			366	9011
				(6430)

New York City $\frac{9011}{366} = 24$ μμc Sr^{90}/g Ca

San Francisco $\frac{6430}{366} = 17$ μμc Sr^{90}/g Ca

DISCUSSION

Dr. S. Allan Lough, Chairman of the session, directed the discussion which follows.

LOUGH: I am confident that all of you have reflected on the problem of predictions. What the fission yield is from the recently tested weapons in my mind is completely unresolved. I do not know. And I would like to ask any of these three gentlemen, particularly Mr. RIVERA and Dr. KNAPP, to discuss this part of their considerations.*

RIVERA: I don't have any inside information. But, according to estimates in the newspaper articles, there have been 55 MT of fission equivalent injected in the recent Soviet test series, which comes about from an assumption of a 50% fission yield from a total energy yield of 110 MT. That is the figure Bill COLLINS used in his extrapolations, and it is the one I used, too.

KNAPP: Unfortunately, I may have some inside information, and I want to point out I was very careful to talk only about what would happen if a 100 MT weapon deriving 50% of its yield from fission were exploded, under such conditions that its fallout were deposited in the same manner as in the U.S.S.R. in the fall of 1958. A Washington Post article about these results noted that 100 MT is just about equal to the total yield of the Russian test series and that the table I have given can be used to predict milk levels next spring. But everything depends on what the fission yield was, and I don't think anyone can be very sure of that yet.

HOLLAND: Along another line, I don't know whether others share my confusion, but I would like to ask Dr. GRUMMIT how he uses the terms "mean" and "average". This seems to differ from what I am accustomed to.

GRUMMITT: Others share your confusion. I do, too. It does differ from what I normally use. The word should have been "mode". It is the value at the peak, that is, at the maximum, as against the mean or average, and I suppose it was my hurry that caused me not to stop and take time to explain it.

KULP: I wish to comment on analysis of the contribution of the rate and cumulative factors to the Sr^{90} concentration in milk that Dr. KNAPP presented. We have approached this in a slightly different way, which I think reduces some of the uncertainty which exists if the coefficients are computed from the data of individual months. Moreover, I don't think you should take all of North America, because you are combining very different rainfall areas. We computed similar coefficients for two separate rainfall areas; the 40-60 in. rainfall zone, which is most of the eastern U. S., east of the Mississippi and the 30 to 40 in. rainfall zone. In these two zones there are a sizable number of Dr. ALEXANDER's soil stations. There are also a fair number of rain sampling stations in these particular zones, and many milk stations. In addition to the Public Health Service network, our calculation was based on all of the milk data including the extensive LASL powdered milk network. All of these data were used to obtain the average concentration of Sr^{90} in milk and the total deposition in $mc/mi.^2$ for the 6 month growing season, April 1 - October 1, and the cumulative deposit at the midpoint of the growing season, taken as July 1. The coefficients for the rate and cumulative factors are then calculated using the simultaneous linear equations for 1959 and 1960. The best solution uses the years 1959 and 1960 since in the earlier years comulative deposition and the fallout rate were increasing together, so that your difference terms are large compared to the uncertainties. In 1959 and 1960 the fallout rate dropped by a factor of 3 to 5, and the cumulative deposition was essentially the same. The coefficients which we derived for this are about 0.6 for the rate and about 0.12 for the cumulative. If these are used to predict the levels in 1957, 1958 and 1961, there is good agreement with observed levels. Using these coefficients the calculated fraction of the Sr^{90} in milk to the rate factor is about 55%. This may be compared with (R. Scott) Russell's earlier prediction of 80% for England or Wales (which with more recent work they have reduced to 60-70%).

RIVERA: I would like to say that one of the things which strikes me is that these various approaches seem to be in fairly good agreement for the cumulative factor. And, of course, the cumulative factor is going to be the really important one, because that will tell us what the Sr^{90} content of the diet is going to be that we are going to have to live with it for a long time.

ALEXANDER: I think that there are two things to bear in mind here. One is that unlike things should not be put together in an average, and the other is that information should not be thrown away. San Francisco and New York should not agree. Grazing time, for instance, is 6 months out of phase on the West Coast from that on the East Coast. The same applies with respect to a monthly basis of any kind. As for the Midwest, there is no grazing of any kind in the winter months. I think that there is a real hazard in throwing together things that are diametrically opposed in making an overall potpourri.

BRAR: Radioactive fallout from current Russian tests was originally detected at Chicago by us around the 15th of September. On October 4th we collected food samples from Food-Chain Stores for gamma-ray activity measurements. A sample of spinach (washed and packaged) showed considerable amount of Ba^{140}-La^{140} radioactivity. This kind of situation illustrates how radioactivity from different parts of the country can be transported around in such a short time. This, of course, can easily throw off the "local" radioactivity measurements by an unknown factor.

LOUGH: I don't think there is anyone who is not aware that these efforts at generalizations involve tremendous uncertainties because of the paucity of data and the difficulty of getting samples and enough time to analyze these samples, both with respect to geographic distribution and with respect to time, not to mention the various things that should be sampled. And then, there are the uncertainties as to what people eat. So, we have a lot of variables in here, and all we are trying to do is to get the best estimate we can under the conditions. But I don't think that we should forget that it is really a guess, and high powered mathematics will lead to results, but we don't know whether the results come from very reliable sources.

HOLLAND: Since Dr. LANGHAM has only the population in his title, I take the liberty of referring to the Los Alamos Scientific Laboratory data on Cs^{137} in milk. As I am sure you know, ANDERSON AND LANGHAM have correlated the levels in milk with precipitation several times and found rather different regimes in different parts of the country. I spent some time this summer at Los Alamos studying this and actually changed the independent variable to fallout rate, as measured by the Sr^{90} collections. You do have considerable regional variations in the ratio of Cs^{137}/K in milk to the fallout rate, suitably time-weighted; so that I have no reason to doubt this would also be true of the Sr if it were examined on a regional basis.

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*Since announcements have been made after November 17, 1961 giving an estimated fission yield of about 25 MT for the 1961 USSR test series, papers giving predictions have been changed by the authors reflecting this knowledge.

AN INVESTIGATION OF THE REPRESENTATIVENESS OF 'GRAB-SAMPLING' IN A SR^{90} IN DRIED MILK PROGRAM PRELIMINARY RESULTS

P.M. Bird and P.G. Mar
Radiation Protection Division
and
G.H. Josie and F.E. Hobson
Research and Statistics Division
Department of National Health and Welfare
Ottawa, Canada

Introduction

The routine analysis for Sr^{90} in milk powder samples has formed part of a nation-wide radioactive fallout monitoring program operated by the Canadian Department of National Health and Welfare since 1955. The primary purpose of the entire program is to estimate the current and future significance to the health of Canadians of radioactive fallout resulting from nuclear explosions.

It is well-known that for "western" diets the Sr^{90}/g Ca levels in milk powder are closely similar to the overall Sr^{90} level in the average diet. It is, therefore, desirable to have some knowledge of the representativeness of the Sr^{90} in milk assay program from the point of view of the dietary intake of milk and dairy products. The representativeness of a sampling program can be assured initially by careful and thorough planning, or it can be evaluated subsequently by special studies superimposed upon an initial arbitrary sampling program.

A thorough study is dependent upon a number of factors including having the necessary production and consumption data for milk, good dietary statistics both for the country and for regions, and

available staff to evaluate the data and recommend the sampling and analysis program. However, against this must be considered the large uncertainties in the biological significance of a given radiation dose, the limitations to the dietary data resulting from either the method of dietary data collection or the smoothing out of possible regional differences due to widespread distribution of some food products, and finally, the necessity for the practical development of a monitoring program quickly.

This paper will outline briefly the purpose and preliminary results of a special study, superimposed upon a regular arbitrary milk sampling program, designed to shed some light on the representativeness of the "grap-sampling" techniques used in the regular program. Representativeness here refers to location and time, and not to dietary intake.

Objectives of the Special Study

The nation-wide program involves some 13 milk powdering plants and once-a-month collection of milk powder (on the 15th or nearest convenient day). An examination of the results obtained from this network of stations over several years would lead one to suspect that there are significant differences between different regions in Canada. There is however, no evidence to indicate how representative the result for a given station may be for its region nor for the month in which the sample was collected.

This study therefore had four objectives:

1. To investigate the variations between:

    (a) determinations (within days)
    (b) consecutive days
    (c) non-consecutive days within one month
    (d) towns
    (e) classes of towns

2. To answer the two key questions concerning the use of a composite:

    (a) how representative of the composite is one determination?
    (b) how representative of the total production is the composite?

3. To relate the foregoing estimates and analyses to the experimental (laboratory) error as indicated by duplicate analyses.

Thus Winchester and Kemptville were selected as representative of the "lowland" section (Grazing Class I) and Eganville and Shawville as representative of the "highland" section (Grazing Class II). The 6 other towns selected for sampling were Perth, Gananoque, Almonte, Athelstan, Cobden and Napanee, with Napanee peculiar to itself in that its milk-shed is very high in lime compared with the others.

The sampling plan called for a total of 510 lbs. of milk powder collected from the 10 towns as follows:

For the 6 towns (Perth, Gananoque, Almonte, Athelstan, Cobden and Napanee) two 1-lb. samples were to be collected for each of 5 days (Monday - Friday) during 4 consecurive weeks, one at 10 a.m. and one at 2 p.m. - a total of 240 1-lb. samples.

For the 4 towns (Winchester, Kemptville, Eganville and Shawville) identical sampling as above plus double samples (2 lb.) morning and afternoon on 5 days selected randomly during the 20 sampling days. In addition, for these 4 towns, a further series of 1-lb. samples collected morning and afternoon for the first 5 days of sampling - a total of 270 1-lb. samples for these 4 towns. The specified daily collection times (10 a.m. and 2 p.m.) were recommended as being most practical and because the mechanics of the fluid milk delivery routes and the powdering process were such that the samples could be termed random samples. In practice the samples were in fact not collected at the specified times, in some cases 2 were made in the same half-day. However this strengthens, rather than weakens, the case for considering them to be random samples.

The sampling plan is detailed in Table 1. Sampling was undertaken for 4 consecutive weeks beginning June 22, 1959.

Outline of Sequential Analysis Plan

As indicated above it had been decided to analyze the samples according to a step-by-step plan to make the maximum use of the 100 analyses limitation. The actual detailed plan was broken down into the following schedule of stages:

TABLE 1

Schedule for Collection of Dried Milk Samples
No. of 1-lb. Samples to be Collected at Places and Time Specified (1)

(2) Day	1 Winchester		2 Kemptville		3 Shawville		4 Eganville		5 Perth		6 Napanee		7 Gananoque		8 Cobden		9 Almonte		10 Athelstan	
	10 a.m.	2 p.m.	10 a.m.	2 p.m.	10 a.m.	2 p.m.	10 a.m.	2 p.m.	10 a.m.	2 p.m.	10 a.m.	2 p.m.	10 a.m.	2 p.m.	10 a.m.	2 p.m.	10 a.m.	2 p.m.	10 a.m.	2 p.m.
1	3	3	2	2	2	2	2	2	1	1	1	1	1	1	1	1	1	1	1	1
2	2	2	3	3	2	2	2	2	1	1	1	1	1	1	1	1	1	1	1	1
3	2	2	2	2	2	2	3	3	1	1	1	1	1	1	1	1	1	1	1	1
4	2	2	2	2	3	3	2	2	1	1	1	1	1	1	1	1	1	1	1	1
5	2	2	2	2	2	2	3	3	1	1	1	1	1	1	1	1	1	1	1	1
6	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
7	1	1	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1
8	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
9	1	1	1	1	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1
10	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
11	1	1	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1
12	1	1	1	1	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1
13	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
14	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
15	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
16	1	1	1	1	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1
17	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
18	1	1	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1
19	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
20	1	1	1	1	3	3	1	1	1	1	1	1	1	1	1	1	1	1	1	1
Sub-Totals	34	34	34	34	34	34	33	33	20	20	20	20	20	20	20	20	20	20	20	20
Totals 510	68		68		68		66		40		40		40		40		40		40	

(1) Each sample must be accurately and clearly labelled as to: (a) Location (b) Time - month, day, hour (c) Sample No. when more than one sample at same hour.

(2) Dates, as well as day number, can be shown when study date is determined. Days are five consecutive days, Monday - Friday of four consecutive weeks.

(a) 40 Sr^{90} determinations for the 4 towns (Winchester, Kemptville, Eganville and Shawville) on the 5 randomly selected days; using 40 milk powder samples. The statistical analysis of these results would be directed to show whether or not there are significant differences between grazing classes, between towns within a class, between days within towns within classes, and between determinations within days within towns within classes.

(b) 12 Sr^{90} determinations for the 4 towns on the monthly composite including one replicate with duplicate determinations; using 160 samples. The statistical analysis would be directed to show whether or not there are significant differences between classes and between towns within a class. It would also yield a measure of laboratory error and would permit a comparison of monthly estimates from this stage with those derived from (a).

(c) 12 Sr^{90} determinations for the 6 towns (Perth, Gananoque, Almonte, Athelstan, Cobden and Napanee) on the monthly composite; using 240 samples. In combination with (b) this represents a study of monthly composites for all 10 towns. The statistical analysis would be directed to show whether or not there are significant differences between towns.

(d) 8 Sr^{90} determinations for the 4 towns on the weekly composite made up for the 5 randomly selected days; using 40 samples. The statistical analysis would be directed to test the representativeness of the composite as a single determination compared with a weekly value derived from the individual days.

(e) 30 Sr^{90} determinations for the 4 towns on 5 consecutive days; using 30 samples. The statistical analysis would be directed to show whether or not there are significant differences between consecutive days.

Results

To date it has been possible to complete only the first group of 40 Sr^{90} determinations. Even in this case 2 results for Winchester are missing due to lost samples. The Sr^{90} results are given in Table 2.

The statistical analysis of these results has been carried out as shown in Tables 3 and 4. These results can be summarized as follows:

(a) There is no statistically significant difference in the mean Sr^{90} estimates between the "grazing" classes.

TABLE 2

Sr^{90} Determinations in Selected Milk Powder Samples

Winchester			Kemptville			Shawville			Eganville		
Date	Collection Time	Sr^{90} μμc/g Ca	Date	Collection Time	Sr^{90} μμc/g Ca	Date	Collection Time	Sr^{90} μμc/g Ca	Date	Collection Time	Sr^{90} μμc/g Ca
June 22	9:30 a.m.	15.35	June 23	9:00 a.m.	12.77	June 25	10:30 a.m.	15.84	June 24	11:00 a.m.	12.68
	3:00 p.m.	11.60		11:30 a.m.	12.44		2:15 p.m.	14.95		5:00 p.m.	14.19
July 1	8:00 a.m.	11.94	June 29	10:00 a.m.	14.50	June 30	11:35 a.m.	13.53	June 26	11:00 a.m.	15.48
	3:00 p.m.	8.68		2:00 p.m.	13.79		2:05 p.m.	14.73		5:00 p.m.	12.09
July 9	11:00 a.m.	10.84	July 8	9:15 a.m.	11.69	July 6	11:30 a.m.	14.70	July 2	11:00 a.m.	12.77
	3:00 p.m.	11.08		10:30 a.m.	12.13		2:15 p.m.	13.00		5:00 p.m.	9.35
July 14	9:00 a.m.	10.14	July 10	9:20 a.m.	13.60	July 15	11:35 a.m.	16.15	July 7	11:00 a.m.	10.77
	3:00 p.m.	9.76		10:15 a.m.	12.36		2:05 p.m.	14.97		4:00 p.m.	10.17
			July 16	9:30 a.m.	11.86	July 17	10:30 a.m.	17.80	July 13	11:00 a.m.	12.21
				10:45 a.m.	13.28		2:00 p.m.	16.61		5:00 p.m.	11.28

TABLE 3

Sr^{90} Estimates - Analysis of Results
($\mu\mu c/g$ Ca)

Class	Town	Day	A.M. Rep	P. M. Rep II	Sum	Mean	AM-PM Diff.
I	Winchester	June 22	15.35	11.60	26.95	13.4750	3.75
		July 1	11.94	8.68	20.62	10.3100	3.26
		July 9	10.84	11.08	21.92	10.9600	-0.24
		July 14	10.14	9.76	19.90	9.9500	0.38
		-	-	-	-	-	-
		Sum	48.27	41.12	89.39		
		Mean	12.0675	10.2800		11.1738	
	Kemptville	June 23	12.77	12.44*	25.21	12.6050	0.33
		June 29	14.50	13.79	28.29	14.1450	0.71
		July 8	11.69	12.13*	23.82	11.9100	-0.44
		July 10	13.60	12.36*	25.96	12.9800	1.24
		July 16	11.86	13.28*	25.14	12.5700	-1.42
		Sum	64.42	64.00	128.42		
		Mean	12.8840	12.8000		12.8420	
	I	Sum	112.69	105.12	217.81		
		Mean	12.5211	11.6800		12.1006	
II	Shawville	June 25	15.84	14.95	30.79	15.3950	0.89
		June 30	13.53	14.73	28.26	14.1300	-1.20
		July 6	14.70	13.00	27.70	13.8500	1.70
		July 15	16.15	14.97	31.12	15.5600	1.18
		July 17	17.80	16.61	34.41	17.2050	1.19
		Sum	78.02	74.26	152.28		
		Mean	15.6040	14.8520		15.2280	
	Eganville	June 24	12.68	14.19	26.87	13.4350	-1.51
		June 26	15.48	12.09	27.57	13.7850	3.39
		July 2	12.77	9.35	22.12	11.0600	3.42
		July 7	10.77	10.17	20.94	10.4700	0.60
		July 13	12.21	11.28	23.49	11.7450	0.93
		Sum	63.91	57.08	120.99		
		Mean	12.7820	11.4160		12.0990	
	II	Sum	141.92	131.34	273.27		
		Mean	14.1930	13.1340		13.6635	

* A.M. readings

$$\text{Overall Mean} = \frac{217.81 + 273.27}{18 + 20} = \frac{491.08}{38} = 12.9232$$

Rep I (A.M.) Mean 13.4011
Rep II (P.M.) Mean 12.4453
Difference 0.9558

TABLE 4

Summary of Statistical Analysis

$X_{ijkm} = \mu + a_i + \beta_{ij} + \gamma_{ijk} + \delta_{ijkm}$

where i = 1, ... a Classes
j = 1, ... b = n_i Towns Within Classes
k = 1, ... c = n_{ij} Days within Towns
m = 1, ... d = n_{ijk} Determinations within Days

All effects, except μ, are considered N I D (o, σ_e^2) where $_e^2 = {}_a^2 \; {}_b^2, {}_c^2, {}_d^2$, respectively.
Universe: 100 mi. radius of Ottawa.
Class 1 : lowland open plain with good grasses and clovers.
Class 2 : grazing area running up into the pre-Cambrian where the poorer grasses grow.

Analysis of Variance

Source of Variation	d.f.	S.S.	M.S.	E (M.S.)
Classes (A)	1	23.1422	23.1422	$\sigma d^2 + 2\sigma_c^2 + 9.5322\sigma_b^2 + 18.9474\ {}_a^2$
Towns (B) in A	2	61.3224	30.6612	$\sigma d^2 + 2\sigma_c^2 + 9.4444\sigma_b^2$
Days (C) in B	15	51.8756	3.4584	$\sigma d^2 + 2\sigma_c^2$
Det. (D) in C	19	31.9384	1.6810	σd^2
Total	37	168.2786		

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$H_{o,1}$: $\sigma_c^2 = 0$, i.e. there is no sig. diff. btwn Days within Towns. $F_{15,19} = 3.4584/1.6810 = 2.06$. Do not reject $H_{o,1}$

$H_{o,2}$: $\sigma_b^2 = 0$, i.e. there is no sig. diff. btwn Days within Classes $F_{2,15} = 30.6612/3.4584 = 8.87_{**}$ Reject $H_{o,2}$

$H_{o,3}$: $\sigma_a^2 = 0$, i.e. there is no sig. diff. btwn Classes. Do not reject $H_{o,3}$.

$\bar{x} = 12.9232$ = unbiased estimate of the mean of 100 mile radius of Ottawa.

$$\text{var}(\bar{x}) = \frac{d^2}{38} + \frac{c^2}{19} + \frac{b^2}{4} + \frac{a^2}{2} = \frac{1.6810}{38} + \frac{0.8887}{19} + \frac{2.8635}{4} + \frac{0}{2} = 0.8069$$

Re optimum allocation of resources: decrease the number of days; increase the number of towns; increase the number of Determinations per Day. Because Towns MS is estimated with only 2 d.f. it should be treated with caution.

(b) There is no statistically significant difference in the mean Sr^{90} estimates between days for the same town.

(c) A statistically significant variation was observed between towns.

(d) The a.m. readings are higher than the p.m. readings most of the time (14 out of 19) but the difference between the means (a.m. and p.m.) is not statistically significant.

(e) The maximum difference between two determinations was

$$17.80 - 8.68 = 9.12$$

The maximum difference for any one town (Winchester) was

$$15.35 - 8.68 = 6.67$$

The maximum difference for any one town for one day (Winchester) was

$$15.35 - 11.60 = 3.75.$$

Summary and Conclusions

In evaluating the results obtained in this study, with a view to drawing conclusions which may be applicable to the overall milk sampling program, it is well to keep in mind the purpose of the study and the circumstances under which samples were collected. For instance, it would not be reasonable to assume that the pattern of variability observed for these stations would hold for stations in another part of Canada or that the pattern would hold for the same stations at another time of year. In Canada winter and summer grazing habits are substantially different and one would expect a similar study conducted during the winter to show a different pattern.

In addition, there is now little doubt that the rate at which fallout is deposited on the ground as well as the total amount deposited affect the levels observed in growing plants and grasses. Sampling for this study was conducted at just about the time when the maximum Sr^{90} levels were being observed in milk and it is reasonable to expect that the contribution from fallout rate to those levels was at or near the maximum. It is not possible to predict how the pattern of variability would be changed under conditions when total deposit was the main contributor to the observed levels.

From these comments it is clear that interpretations of the results obtained in this study, as well as those obtained in the nation-wide sampling program, should be made with caution. It is too early yet to draw firm conclusions from the first stage results, but there is an indication that for a nation-wide program a more efficient allocation of a given number of samples would result from an increase in the number of stations in preference to the number of samples from a station. In any event, the results warrant continuation of this investigation according to the scheduled plan.

ACCRETION AND REPLACEMENT OF SKELETAL MINERAL DEDUCED FROM SURVEY IN THE UNITED KINGDOM OF BONES FOR SR^{90}

John F. Loutit, Director
Radiobiological Research Unit
Medical Research Council
Harwell, England

Dr. Loutit summarized some of the results of his recent studies which are now published elsewhere or will be soon. These published documents are listed below for reference. The material summarized pertained to the growth and turnover of the skeleton with particular emphasis on strontium. (Ed.)

Bryant, F. J., and J. F. Loutit. Human bone metabolism deduced from strontium assays. United Kingdom Atomic Energy Authority, Atomic Energy Research Establishment, Harwell, England, report AERE-R-3718, Her Majesty's Stationery Office, London, 1961. 52 pp. (8 shillings).

Loutit, J. F. Lectures given at the University of Chicago, 1961, to be published in: Irradiation of Mice and Men, University of Chicago Press, 1962.

FALLOUT AND THE FOOD CHAIN: A STATUS REVIEW

C. L. Comar, R. H. Wasserman, F. W. Lengemann and J. C. Thompson, Jr.
Department of Physical Biology
Cornell University
Ithaca, N. Y.

I. General Approach

The behavior and metabolism of strontium, cesium and iodine, relevant to the problem of fallout, have been detailed in published review articles (1, 2, 3, 4). It is the intention here to discuss briefly a general approach and then to deal with a limited number of specific subject areas from the standpoint of present status and foreseeable problems.

In the broadest sense the "fallout problem" requires knowledge of the relationships between the radionuclides released to the biosphere and the harm that may thereby be caused to the human population. Figure 1 presents several stepwise categories of investigational approach. The most direct approach is normally to be preferred, which in the present instance would be a study of the relationships between the radionuclides disseminated, and harm to man. However, it is obviously most difficult, if not impossible, to evaluate harm directly primarily because of the relatively low levels of radiation exposure, low dose rates, and long latent periods. Evaluation of harm as a function of known radiation dose from fallout is occupying (as yet without success) the thoughts and efforts of our best minds, and is seemingly unapproachable by direct frontal investigation.

The problem of radiation dose (a step removed) is not an easy one, but is more susceptible to the experimental approach than is that of harm: the radiation dosage from the bone-seekers is complicated because of heterogeneous distribution; for example, even bone formed throughout life from a diet of constant radiostrontium to calcium ratio may not

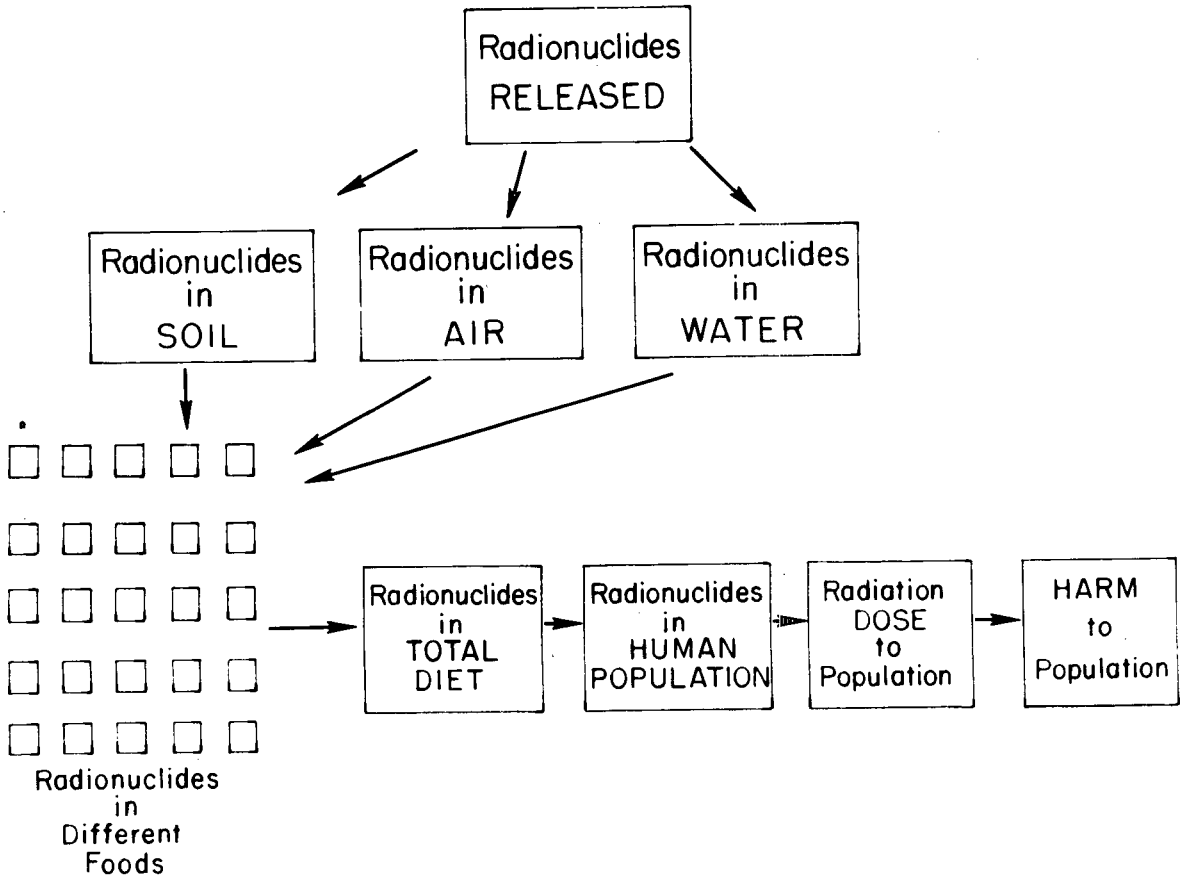

FIGURE 1. Schematic Indication of Step-wise Investigational Approach to the "Fallout Problem".

have a completely uniform distribution. There is, however, a reasonable theoretical and experimental basis for estimation of dose delivered to the gastrointestinal tract from material passing through, and to tissues and organs from radionuclides such as Cs^{137} and I^{131} (5, 6).

It is then of advantage to utilize a still more indirect step, namely the relationship between the radionuclides released and the amount and distribution that will develop in the population. It is pointed out, however, that this relationship is complex because of varying pathways through the food chain, differing dependence on fallout rate and cumulative totals, and differing holdup times. The amounts of radioactivity in man can be measured directly by analysis of human bones for Sr^{90}, by analysis of thyroid glands for I^{131}, and by whole body counting for Cs^{137}. Some of the practical difficulties may be mentioned. Post-mortem samples that are representative of the population are difficult to obtain on a current and continuing basis; for Cs^{137}, post-mortem samples are not required, but the limited number and non-portability of whole body counters restricts sampling to individuals who live in or visit a given area. Results of bone analyses for Sr^{90} in adults are most difficult to interpret because of the heterogeneity of radionuclide deposition; fortunately, values are meaningful for young children who have a reasonably uniform distribution of Sr^{90} throughout the skeleton and who are close to equilibrium with their dietary intake. Also values for the stillborn can be correlated with the maternal diet.

Studies of intake in the diet (a step further removed) can overcome the problem of non-representativeness of post-mortem samples, but introduce another uncertainty, namely the relationship between intake and body burden attained. Some additional advantages of studies on either total diet or individual food items are: (a) unlimited numbers and amounts of samples can be obtained; (b) changes in levels in dietary intake can be observed with only a small time lag; (c) correlations between amounts and rates of fallout and levels in the diet can be made in terms of specific meteorological and agricultural conditions; (d) if specific foods are assayed, the chief contributors of radioactivity can be determined; and (e) the general knowledge obtained can serve as a basis for remedial modifications or controls, and further will permit predictions as to future levels as a function of amount and conditions of future releases. It is emphasized that the usefulness of survey data may depend to a large extent upon the planning and execution of surveys.

Recent experimental work has suggested that urine may be a useful integrating sample for monitoring the dietary intake of Sr^{90} (7,8) and of Cs^{137} (9). The advantages presented for Sr^{90} are as follows: (a) the urinary samples reflect the total amount of Sr^{90} and calcium that enters body tissues, and there is no need to guess the contributions of given food items or the availability of the Sr^{90} in given food items; (b) values are calculated in terms of total intake of Sr^{90} and calcium which is probably the most important parameter governing the body burden; (c) any sudden changes in levels of Sr^{90} would be noted early, and the results could reflect any day-by-day variations; (d) sampling and bulking would be relatively simple and work could be designed to reflect individual intakes as well as group averages classed according to any category of interest; (e) the results could be interpreted to give values for concentrations of Sr^{90} and Sr^{90}/calcium ratios in areas of newly forming and exchangeable bone; and (f) from a continued program of sampling it would be possible to construct the time pattern of radiostrontium levels in bone. The feasibility of large-scale monitoring of urine has not yet been demonstrated.

Information to-date has been, and must continue to be, obtained from work carried on simultaneously in all categories mentioned. It is important in evaluation that the data most suitable for the purpose be given the most weight. Especially must we be cautious of attempts to describe individual and physiological behavior from survey studies; likewise must we avoid extrapolation to populations from experimental studies on limited numbers of individuals. The work can go forward more rapidly and with greater certainty if knowledge at each level can be taken into account in the planning and execution of both surveys and controlled experimental studies.

Adequate information for dealing with fallout must come from firm knowledge of the way specific radionuclides pass through the food chain, and such knowledge will be derived from basic understanding of the biological systems, controlled experimental studies using radiotracers, and studies using environmental radioactivity.

II. Food Surveys

The organization of surveys for radionuclides in foods has recently been given detailed consideration by an expert committee of the Food and Agriculture Organization of the United Nations and a full report is in press. Some brief observations are as follows:

A. Objectives: The design and scope of surveys should be based on the required objectives, some of the more important of which are listed:

(a) Determination of whether or not a given level of radioactivity in the diet is reached;

(b) Estimation of mean dietary radioactivity over appropriate periods of time and of maximum levels in diets of any special groups;

(c) Monitoring of changes in dietary levels;

(d) Studies of the food chain, especially to identify components that are the major contributors of radioactivity, and to clarify the processes and vectors involved;

(e) Studies of correlations between radionuclides released and pathways through the food chain for purposes of prediction;

(f) Studies concerned with effects of modification of diets and of changes in food technology;

(g) Provision of a basis for policy decisions, recommendations or actions by authorities, and provision of information to the public.

B. Acceptable Levels of Radionuclides in Foods: The representativeness of sampling and the precision of assay that is required is determined by the official acceptable levels. An important waste of effort and expenditure may result from attempts to provide accurate evaluation of levels that are small compared to those of significance. Official acceptable levels may be used to limit surveys to the significant specific radionuclides and to the major dietary contributors of the radionuclides.

C. Selection of Foods: For calculation of population exposures, it may be necessary to know the mean intake of certain individual foods, classes of foods (e.g. vegetables, cereals, dairy products), and total intakes. This information is obtainable by (a) nationwide data on food production and disposition, (b) dietary surveys that are inherently limited in scope, and (c) individual consumption surveys that are even more limited.

Considerable information is now available that permits choice for assay of foods that are (a) the major contributors of specific radionuclides, (b) useful for showing time trends because of schedule of harvest or time pattern of contamination, and (c) of importance for special groups such as milk and prepared baby foods that might form a major part of the diet of the very young. Samples are preferably obtained from farms when the interest is mainly in food chain behavior because the history of the sample is often available. For estimation of mean levels, samples are preferably obtained from centers of collection or processing plants, rather than at points of consumption. This usually leads to better data on relationships to conditions of agricultural production, and to a better chance of obtaining widely representative samples.

Assays of total diets are sparing of radiochemical analyses, and are thus of advantage when resources are limited, but it must be remembered that they yield information only on total intakes. Sampling in this instance is most efficiently done in homes or stores.

D. Comparisons of Milk Surveys: In past years various organizations have undertaken independent surveys of differing degrees of sophistication; full descriptions can be found in the literature (10). Some comparisons are attempted here, to indicate the degree of rigor necessary. Following are brief statements of scope and design for programs of milk sampling to be compared.

a. Health and Safety Laboratory Liquid Milk - New York City (HASL-LM): Milk purchased from local stores representing five large dairies.

b. Public Health Service Raw Milk Survey (PHS-RM): Milk collected from a station that represented part of a metropolitan milk-shed and a group of farms milking at least 1000 cows.

c. Public Health Service Processed Milk Survey (PHS-PM): Sample is composite from plants supplying not less than 90% of city's milk supply and taken so as to represent milk consumed in the city.

d. Consumers Union Survey (CU): Samples purchased from local stores in amounts that represent consumption by an individual over a two-week period.

TABLE 1

Comparison of Sr^{90} in Milk Among Various Programs
1958-1961

Time	Programs[1] or Cities				Observations	Significant Difference[2]	
	(Mean value in μμc Sr^{90}/g Ca)					Yes	No
1961							
Jan.-May		PHS-RM	vs	PHS-PM			
	Nine Cities	6.6		5.5	86		x
	Atlanta, Ga.	10.8		6.7	10		x
	Austin, Texas	3.1		2.1	10		x
	Chicago, Ill.	5.8		5.6	8		x
	Cincinnati, Ohio	7.5		6.5	10		x
	New York, N.Y.	6.6		6.6	8		x
	Sacramento, Calif.	3.4		3.7	10		x
	Salt Lake City,Utah	2.9		4.5	10		x
	Spokane, Wash.	6.3		6.8	10		x
	St. Louis, Mo.	13.1		7.1	10		x
Jan.-May		MSDH	vs	PHS-PM			
	Minneapolis, Minn.	7.3		6.8	10		x
January		CU	vs	PHS-RM			
	Four cities	5.3		6.8	8		x
January		CU	vs	PHS-PM			
	Eight cities	5.5		5.4	16		x
1960							
Mar.-Dec.		PHS-RM	vs	PHS-PM			
	Nine cities	7.2		5.6	114		x
	Atlanta, Ga.	11.1		7.8	10	x	
	Austin, Texas	3.0		1.6	12	x	
	Chicago, Ill.	7.4		5.3	12		x
	Cincinnati, Ohio	7.2		6.6	10		x
	New York, N.Y.	7.7		8.1	14		x
	Sacramento, Calif.	2.6		4.3	10		x
	Salt Lake City,Utah	5.4		5.1	20		x
	Spokane, Wash.	9.0		6.7	16		x
	St. Louis, Mo.	12.2		6.9	10	x	
June-Dec.		HASL-LM	vs	PHS-RM			
	New York, N.Y.	7.6		7.7	14		x
June-Dec.		HASL-LM	vs	PHS-PM			
	New York, N.Y.	7.6		8.1	14		x
Aug.Dec.		MSDH	vs	PHS-PM			
	Minneapolis, Minn.	5.6		7.9	10		x
1959							
July-Aug.		CU	vs	PHS-RM			
	Three cities	7.0		13.1	6		x
1958							
July-Aug.		CU	vs	PHS-RM			
	Seven cities	8.0		7.2	14		x

[1] PHS-RM = Public Health Service Raw Milk
PHS-PM = Public Health Service Processed Milk
MSDH = Minnesota State Department of Health
CU = Consumers Union
HASL-LM = Health and Safety Laboratory Liquid Milk

[2] "t" tests were used to determine the existence of significant differences (level of significance = .05)

e. Minnesota State Department of Health (MSDH): Two-ounce samples, collected daily at each of six stations, are composited and analyzed monthly. Collection is made at the bottling machines so that samples are random representations of the milk produced in each milk-shed.

Table 1 shows comparisons between results obtained from the different survey methods which include differences due to analytical as well as collection techniques. It is noted that in only three comparisions are there significant differences. This leads to the tempting suggestion that even the most naive methods of sampling (purchase at a local store) will most likely provide values adequate for assessment of regional population intakes under conditions of widespread fallout. Remember that uncertainties are great in the translation of such levels into harm to the population, and that the levels themselves represent a population exposure that is a fraction of background exposure.

E. Comparisons of Total Diet Surveys: Table 2 presents a comparison of estimates of Sr^{90}/Ca in total diets derived by differing methods of sampling. Again, one is impressed with the degree of agreement.

F. Relative Contributions of Foods to Dietary Sr^{90}: Table 3 presents some data on Sr^{90}/Ca in various constituents as compared to that in the total diet. An important point is that the ratio of total diet to milk has been reasonably constant at about 1.2 to 1.6. This ratio is expected to change with time, approaching 3.7 as an equilibrium value. Meanwhile the observed constancy makes it possible to estimate total diet values from milk data, which are much more numerous. Whole wheat products were less constantly related, as might be expected from known modes of contamination. Table 4 presents values for relative contributions of Sr^{90} from different food classes.

The data underscore another important matter, namely that although milk products were the greatest single contributor of Sr^{90} to the diet, a diet without milk would most likely have had as much as two to three times the Sr^{90}/Ca level.

G. General Comments on Surveys: Information on behavior of radionuclides in the food chain and knowledge of the generalized mode of contamination can be used to simplify food surveys without compromise of objectives.

Areas of needed work are: (1) problems of short-lived radioactivity which may involve studies of short-term flow of food supplies (see Table 5 for study of Northeastern region); (2) delineation of variability; (3) consideration of rural populations where the consumption of milk and leafy vegetables immediately after production might be high; (4) obtaining a continuum of data on specified food products other than milk; (5) investigation of analysis of excretions as integrating samples for the human diet; and (6) satisfaction of the public that appropriate efforts are being taken.

TABLE 2

Comparison of Strontium Units[1] of Various Total Diet Programs in Northeastern United States During 1959

AGENCY-PROGRAM	DATE	LOCATION	μμc Sr^{90}/g Ca
HASL[2] - Total diet estimate from selected foods using USDA consumption data	November	New York, N.Y.	17.7
HASL - Complete diets of five lactating women for five days	May	Boston, Mass.	16.9
HASL - Typical first year infant diet using values reported for processed baby foods in August 1959 and January 1960	August '59 Jan. '60	New York, N. Y.	14.0
HASL - Complete diets of five hospital personnel for six days	December	New York, N. Y.	12.4
Consumers Union - Market purchase of typical teenage diet for two weeks	November	Boston, Mass.	15.5
		Brooklyn, N. Y.	12.2
		Manhattan, N. Y.	12.6
		Pittsburgh	13.2
Aggregate Average			14.3

1 Strontium Unit = μμc Sr^{90}/g Ca
2 Health and Safety Laboratory

TABLE 3

Ratios of Strontium Units[1] in the Total Diet
to Those of Various Food Products in the United States, 1959-1961

Date	Agency	Location	[2]Ratios of Strontium Units in Total Diet to Strontium Units in:					
1961			Milk	Whole Wheat Bread	Whole Wheat Products	All Wheat Products	Vegetables	Fruits and Vegetables
April	HASL[3]	Chicago, Ill.	1.7	.48		.71	.28	.32
"	HASL	San Francisco,Cal.	2.1	.32		.55	.45	.32
Jan.	HASL	New York City	1.2	1.1		.92	.60	.43
"	HASL	San Francisco, Cal.	2.1	2.2		.76	.36	.31
"	CU[4]	Atlanta, Ga.	1.4		.64			
"	CU	Austin, Texas	1.8		.40			
"	CU	Boston, Mass.	1.5		.62			
"	CU	Boulder, Colo.	1.8		.67			
"	CU	Brooklyn, N.Y.	1.1		.16			
"	CU	Chicago, Ill.	1.2		.27			
"	CU	Grand Forks, N.D.	1.9		.58			
"	CU	Louisville, Ky	.88		.41			
"	CU	San Francisco, Cal.	3.6		.77			
"	CU	Seattle, Wash.	1.0		.93			
"	CU	St. Louis, Mo.	1.1		.45			
"	CU	Washington, D.C.	1.8		.29			

TABLE 3 (cont'd)

Date	Agency	Location	Milk	Whole Wheat Bread	Whole Wheat Products	All Wheat Products	Vegetables	Fruits and Vegetables
1960								
Nov.	HASL	Chicago, Ill.	1.2	.45		1.0	.45	.45
Oct.	HASL	New York City	1.7	.48		1.3	.29	.27
Sept.	HASL	Chicago, Ill.	1.4	.44		.68	.36	.47
Aug.	HASL	San Francisco, Cal.	1.7	.58		.50	.51	.49
June	HASL	New York City	1.1	.40		1.1	.41	.49
May	HASL	Chicago, Ill.	1.7	.45		.58	.43	.40
March	HASL	New York City	1.1	.34		.63	.46	.57
March	HASL	San Francisco, Cal.	1.3	1.7		1.1	.39	.54
1959								
Nov.	CU	Atlanta, Ga.	1.4					
"	CU	Boston, Mass.	1.7					
"	CU	Brooklyn, N.Y.	1.1					
"	CU	Louisville, Ky.	.94					
"	CU	New Orleans, La.	1.0					
"	CU	Pittsburg, Pa.	1.1					
"	CU	St. Louis, Mo.	1.3					
"	CU	Washington, D.C.	1.2					
1961	Average		1.6	1.0	.52	.74	.42	.34
1960	Average		1.4	.60	----	0.86	.41	.46
1959	Average		1.2	---	----	---	---	---

[1]Strontium Unit = 1 μμc Sr^{90}/g Ca
[2]Ratios were computed from annual regional food consumption data based on 1955 U.S. Dept. of Agriculture Household Food Consumption Survey
3 Health and Safety Laboratory
4 Consumers Union

TABLE 4

Percentage Contribution of Sr^{90} from Various Food Products to the Annual Sr^{90} Intake in the Total Diet[1]

Date	Agency	Location	Milk	Whole Wheat Bread	All Wheat Products	Vegetables	Fruits and Vegetables
			(Percent of Annual Sr^{90} Intake)				
1961							
April	HASL[2]	Chicago, Ill.	37	2	22	28	40
"	"	San Francisco, Cal.	28	5	24	19	41
Jan.	"	New York City	50	1	16	15	31
"	"	San Francisco, Cal.	29	1	18	24	43
"	CU	Atlanta, Ga.	37				
"	"	Austin, Texas	27				
"	"	Boston, Mass.	39				
"	"	Boulder, Colo.	37				
"	"	Brooklyn, N. Y.	52				
"	"	Chicago, Ill.	53				
"	"	Grand Forks, N.D.	35				
"	"	Louisville, Ky.	56				
"	"	St. Louis, Mo.	60				
"	"	San Francisco, Cal.	18				
"	"	Seattle, Wash.	61				
"	"	Washington, D.C.	27				
1960							
Nov.	HASL	Chicago, Ill.	50	2	15	17	31
Oct.	"	New York, N.Y.	34	2	12	33	50
Sept.	"	Chicago, Ill.	44	3	24	19	25
Aug.	"	San Francisco, Cal.	34	3	27	16	28
June	"	New York, N.Y.	53	3	17	21	27
May	"	Chicago, Ill.	36	2	25	21	35
March	"	New York, N.Y.	50	2	18	22	24
"	"	San Francisco, Cal.	47	1	12	23	30

(Continued)

TABLE 4 (Continued)

Date	Agency	Location	Milk	Whole Wheat Bread	All Wheat Products	Vegetables	Fruits and Vegetables
			(Percent of Annual Sr^{90} Intake)				
1959							
Nov.	CU	Atlanta, Ga.	34				
"	"	Boston, Mass.	34				
"	"	Brooklyn, N.Y.	52				
"	"	Louisville, Ky	52				
"	"	New Orleans, La.	48				
"	"	Pittsburg, Pa.	52				
"	"	St. Louis, Mo.	50				
"	"	Washington, D.C.	41				
"	PHS	[4]Cincinnati, Ohio	37		23		38
"	HASL	New York, N.Y.	39		16		40
May	HASL	Boston, Mass.	44				42
1958	PHS	Cincinnati, Ohio	36		15		42
1958	HASL	New York, N.Y.	40		27		31
1961	Average		41	2	20	22	39
1960	"		44	2	18	22	32
1959	"		44	--	20	--	40
1958	"		38		22		36

1 Annual Regional food consumption data obtained from 1955 U.S. Dept. of Agriculture Household Food Consumption Survey used to compute intake values for HASL TriCity Diet and Consumers Union Diet Studies.
2 Health and Safety Laboratory
3 Consumers Union
4 Public Health Service

TABLE 5

Appropriate Marketing Times for Major Food Products Consumed in Northeastern United States

Food	Days in Marketing Channel: Local Area	Days in Marketing Channel: Distant Area		Maximum Storage Period	Harvest Period[1]: W	Sp	S	F
Dairy Products:								
Butter	6-8	10-15		10-12 months				
Cheese Processed	7-10	20-30		10-12 months				
Cheddar	60-90	70-100		12 months and up				
Cottage Cheese	3-5			10-14 days				
Fluid Milk	3-5			2-3 weeks				
Ice Cream	6-8	10-15		6-12 months				
Processed Milk		10-20		12 months and up				
Vegetables:								
Asparagus	3-5	10-12		2 weeks		x	x	
Broccoli	3-5	10-14		2-3 weeks	x	x		x
Brussel Sprouts	3-5	10-14		2-4 weeks	x			x
Cabbage	3-5	6-10		4-6 months	x	x	x	x
Cantaloupe	3-5	10-14		2-3 weeks		x	x	x
Carrots	3-5	7-10		6-8 months	x	x	x	x
Cauliflower	3-5	10-14		2-3 weeks	x	x	x	x
Celery	3-5	6-10		2-3 weeks	x	x	x	x
Corn	2-4	5-8		2-3 weeks	x	x	x	x
Cucumbers	3-5	6-8		2-3 weeks	x	x	x	x
Escarole	3-5	10-14		4 weeks		x	x	
Green Beans, peas	3-5	6-10		2-3 weeks	x	x	x	x
Lettuce	3-5	10-14		3-4 weeks	x	x	x	x
Onions Fresh	3-5	6-8		2-3 weeks	x	x	x	
Cured	10-14	20-30		6-8 months			x	x
Peppers	3-5	6-8		2-3 weeks	x	x	x	x
Potatoes	3-5	5-14		12 months	x	x	x	
Spinach	3-5	6-8		1-2 weeks	x	x	x	
Squash Summer	3-5	7-10		2-3 weeks	x	x	x	
Winter	3-5	7-10		4-6 months			x	x
Tomatoes	3-5	8-10	Green	4 weeks				
			Ripe	10 days	x	x	x	x
Watermelon	3-5	6-10		3-4 months			x	

(Continued)

TABLE 5 (Cont'd)

Food	Day in Marketing Channel: Local Area	Day in Marketing Channel: Distant Area	Maximum Storage Period		Harvest Period: W	Sp	S	F
Fruits:								
Apples	3-5	10-14	6-12 months				x	x
Oranges		10-14	6-12 months		x	x	x	x
Grapefruit		6-10	6-12 months		x	x	x	x
Apricots		10-14	2-3 weeks				x	
Cherries	3-5	8-10	1-2 weeks				x	
Lemons		10-14	6-12 months		x	x		x
Cranberries		4-6	4-6 weeks					x
Grapes	3-5	10-14	3-4 weeks				x	x
Peaches	3-5	6-8	2-3 weeks				x	
Pears	3-5	10-14	2-6 months				x	x
Plums	3-5	10-14	3-4 weeks				x	x
Strawberries	2-4	6-8	10-14 days			x	x	
Processed Products:								
Canned Vegetables	7-10	15-20	Years					
Canned Fruits	7-10	15-20	Years					
Frozen Vegetables	7-10	15-20	Years					
Frozen Fruits	7-10	15-20	Years					
Frozen Juices	6-8		Years					
Dried Fruits		30-40	12 months					
Dried Vegetables	10-20	30-40	12 months					
Meat:								
Beef	7-10	10-15		Frozen 12 months				
				Fresh 1 month				
Fish	5-10	10-20		Fresh 2-4 weeks				
	10-20	20-30		Canned 12 months and up				
Lamb	5-10	10-15		3-4 weeks				
Pork Cured	10-15	15-20		Frozen 4 months				
				Cured 12 months				
Fresh	2-5	5-10		Fresh 1 month				
Poultry	4-7	6-10		Frozen 12 months				
				Fresh 10 days				
Veal	5-10	10-15		Frozen 12 months				
				Fresh 1 month				
Flour	15-30	30-40	12 months and up					
Eggs	5-7	7-10	2-3 weeks					

W = Winter; Sp= Spring; = S = Summer; F = Fall.

III. Retention of Radiostrontium in Man

Retention of ingested radiostrontium can be judged from two standpoints: (a) absolute retention and (b) comparison with calcium retention by consideration of $OR_{body/diet}$ values ($OR_{body/diet} = \frac{Sr^*/Ca\ body}{Sr^*/Ca\ diet}$). Absolute retention values are difficult to interpret; for example, in a mature individual exposed to Sr^{90}, the daily net retention may be about 20% of the amount ingested per day to start with, but should eventually decrease to zero as the skeleton becomes labeled under constant intake. Under such conditions of constant intake, the Sr^{90} content of the body would be expected to reach a constant value just as the calcium content does.

Evaluation of the $OR_{body/diet}$ in man is of most practical interest in connection with chronic ingestion. Numerous independent studies have established $OR_{body/diet}$ values for a variety of animal species using varying methods based on differing assumptions (2,11,12). It is, however, not possible to obtain needed information from direct observations on man. All of the observations to date have a considerable degree of uncertainty for one methodological reason or another. $OR_{body/diet}$ values of 0.29 were reported from controlled studies using retention of an ingested dose of Sr^{85} and Ca^{45} in sixteen patients (13); earlier measurements using similar procedures gave values of 0.44 and 0.54 for groups of patients on two different types of diet (14). An $OR_{body/diet}$ value of about 0.25 was obtained from 4 normal healthy women by an indirect method, namely by measurement of fallout Sr^{90} and stable calcium to give an $OR_{milk/diet}$ value of 0.1 from which the $OR_{body/diet}$ value was calculated using an $OR_{milk/plasma}$ value of 0.4 as observed in the cow and goat (15).

From studies on calves and rats (17,18), it was suggested that discrimination against strontium in the process of gastrointestinal absorption is reduced in the young. Bryant and Loutit (16) estimate

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Sr^* = isotope of strontium.

confirmed (24). However, in both of these experiments, a quantitative inverse proportionality between the calcium in diet and Sr^{90} in milk was not seen although the $OR_{milk/diet}$ was nevertheless independent of calcium intake. On theoretical grounds it was therefore predicted that, with time, a quantitative inverse relationship would occur. Since both of these studies were short-term and it was shown that several months are necessary for cattle to adapt to changes in dietary calcium (4), a long-term investigation was undertaken to study this point. In this experiment, 10 weanling pure-bred Holstein heifers were divided into two groups; one group received a medium calcium diet, and the other a high-calcium diet. These were fed through their growing period, through gestation and into lactation. The calcium/phosphorus ratios of the feeds of the two groups were about the same and within the normal range.

About two months after parturition, a balance experiment was performed, using accepted nutritional procedures. Each animal received a known amount of Ca^{47} and Sr^{85} by capsule twice daily for 10 days, and the

TABLE 6

Long Term Effect of Dietary Calcium Level on Relative Secretation of Calcium and Strontium into Milk as Determined by Three Methods

	High Calcium	Medium Calcium
No. of cows	5	4
Ca intake (g /day)	92	44
Sr intake (mg/day)	180	124
Diet (mg Sr/ g Ca)	1.95	2.80
Milk (mgSr/g Ca)	0.19 ± 0.03*	0.23 ± 0.03
$OR_{milk/diet}$	0.098 ± 0.017	0.083 ± 0.009
Ca^{47} (% daily dose/liter of milk)	0.57 ± 0.04	1.04 ± 0.06
Sr^{85} (% daily dose/liter of milk)	0.047 ± 0.005	0.091 ± 0.008
$OR_{milk/diet}$	0.084 ± 0.004	0.089 ± 0.009
Diet ($\mu\mu c\ Sr^{90}$/ g Ca)	32.8	69.9
Milk ($\mu\mu c\ Sr^{90}$/g Ca)	4.1 ± 0.4	8.0 ± 1.1
$OR_{milk/diet}$	0.12 ± 0.01	0.11 ± 0.02

* Standard error of mean

an $OR_{body/diet}$ of about 0.5 for young infants. Preliminary data from two independent studies in the United States, using balance methods for dietary and excreted Sr^{90} and calcium in young infants, suggest that the $OR_{body/diet}$ may be even higher.

Attention is called to a series of three notes that deal with observations on the effects of breast feeding versus feeding with cow's milk (19,20,21). Further data are needed for clarification on points raised which suggest an increased strontium retention in babies fed cow's milk because of an increase phosphate intake as compared with that from breast milk.

Whether or not the higher $OR_{body/diet}$ value in the young is of importance in governing the hazard, depends upon the mechanisms of injury production. If the major factor is the integrated dose over the years, then the higher OR value may not be significant because of the high rate of bone turnover in the young, which will lead to a body burden regulated by the OR at ages of 1 to 2 years and above. On the other hand, if injury is primarily related to an initial insult, then the high OR in the very young may need to be taken into account.

An important area of investigation would be study of the change in discrimination that occurs in the early years of life.

IV. Transference of Radiostrontium into Milk

An important interest in the metabolism of Sr^{90} in animals is in the extent to which it may be transferred into milk.

It is known that the amount of ingested calcium secreted into milk is directly correlated with the total amount of milk and therefore of milk calcium secreted; because of $OR_{milk/diet}$ is roughly constant, it would be expected that the total Sr^{90} secreted into milk would likewise be related to milk yield. Thus it seems more useful to discuss amounts of Sr^{90} in terms of percentage of ingested dose per liter of milk or per gram of milk calcium, since this tends to compensate for differences in milk production when data are to be compared. Values ranging from 0.5 to 2% of an ingested dose of Sr^{*} in the total milk of the cow have been reported (4,22,23). In comparison, with continuous ingestion under normal conditions of feeding, several groups have found about 0.08% of the daily dose to be secreted per liter of milk (24,25,26).

It is of interest to note that the percent of ingested radiostrontium secreted per liter of milk is more than ten times higher for the goat than for the cow. This is because the goat puts a much larger proportion of its dietary calcium into the milk per liter than does the cow.

Values of $OR_{milk/diet}$ are more useful for many purposes than are absolute quantities. Values for the cow and goat have been reported from 0.05 to 0.16, but the majority of more recent experimental measurements fall close to 0.11 (4,12,22,24,26,27). The physiological processes which contribute to this ten-fold differential in calcium and strontium secretion into milk were initially determined in the goat (27), and confirmed in subsequent studies in the cow (22, 24). For clarity of expression, the contribution of any process to the overall discrimination (as given by the OR has been termed the "discrimination factor" (DF). The discrimination factors that result in the $OR_{milk/diet}$ of about 0.1, are 0.2-0.3 for gastrointestinal absorption, 0.6-0.8 for urinary excretion, and 0.5 to 0.6 for mammary secretion. It is evident that the preferential absorption of calcium from the intestinal tract is the major process contributing to the $OR_{milk/diet}$ of 0.1. In experiments relating to the mechanism of mammary discrimination, Twardock, Prinz and Comar (28) assessed the comparative binding of calcium and strontium by milk proteins and found that milk proteins bound strontium to a greater degree than calcium. Since this is the opposite of the action necessary to cause preferential calcium movement into milk, it was concluded that some factor other than protein binding was responsible.

The differential movement of Sr^{90} and calcium in lactating women was recently determined by Lough, Hamada and Comar (29). Like the cow and goat, the $OR_{milk/diet}$ in 4 of 5 women ranged from 0.085 to 0.13, with an average of 0.10; the fifth individual had subnormal intake of calcium, which most likely invalidated the experimental method.

A question of potential practical importance concerns the effectiveness of high levels of dietary calcium in decreasing the level of Sr^{90} in milk. A short-term study on this problem several years ago suggested that increasing the calcium intake of dairy cows would, in fact, decrease the amount of Sr^{90} per liter of milk (4); in a more recent experiment of a short-term nature, these findings were

quantity of the ingested radionuclides transferred to milk was determined. Before radionuclides were administered, samples of milk from each cow and samples of their contemporary feed, were sent to the Health and Safety Laboratory of the New York Operations Office, U.S. Atomic Energy Commission, for fallout Sr^{90} and stable strontium analysis. These data have been summarized and are given in Table 6. These different methods again showed that the $OR_{milk/diet}$ is very predictable and varies from 0.083 to 0.12, depending on technique. Within each method, the $OR_{milk/diet}$ was independent of level of calcium in the diet. As had been predicted, a proportional inverse relationship was found to exist between calcium intake and concentration of Sr* in milk. In the double tracer study with Ca^{47} and Sr^{85}, a 2.2-fold increase in Ca intake decreased Sr^{85} secretion into milk by a factor of about 1.9; in these same animals, the Sr^{90} level in milk was reduced similarly by a factor of 2.0; thus, these data were mutually confirmatory.

The effect of stable strontium on radiostrontium secretion into milk was also investigated in a short-term study (12). In these experiments, two goats were fed a diet containing about 3.0% strontium and 0.25% calcium; the control diet for reference purposes contained 1.7% calcium. Each diet, on an equivalence basis, contained about the same amount of total alkaline earths. After consuming the diet over a preliminary period of two weeks, the goats were given Ca^{45} and Sr^{85} twice daily for 10 days. At steady state, the high-strontium goats and the high-calcium goats secreted 0.77 and 0.74% of the daily dose of Sr^{85} per liter of milk, respectively. The $OR_{milk/diet}$ was about the same for both treatments and was found to be 0.12. Thus, there appeared to be little advantage in using stable strontium as a diluent for radiostrontium, even though the stable strontium supplement represented an increase in total strontium by many orders of magnitude. Also, prolonged feeding of stable strontium in itself would undoubtedly prove deleterious to the health of the animal (30,31). In the similar experiment with two cows, stable strontium had little effect on Sr^{85} secretion into milk.

In recent studies, other feeding procedures were attempted to reduce the transfer of radiostrontium from diet to milk. High levels of potassium phosphate (K_2HPO_4), sodium ethylenediaminetetraacetate (NaEDTA), ion exchange resin (Dowex 50) and magnesium sulfate ($MgSO_4$) were incorporated in the grain mixture of lactating cattle. It was observed that, on a short-term basis, none of these dietary additives decreased the level of Sr^{85} per liter of milk. In fact, EDTA feeding had the opposite effect of increasing Sr^{85} concentrations in milk; these results paralleled earlier observations made with EDTA in the rat (32).

V. Radioiodine in Animal Products

Radioiodine reaches the human population primarily by means of milk or milk products. Dairy animals in foraging necessarily graze over large surface areas, and in doing so can, under some circumstances, produce milk that contains significant amounts of I^{131} even though the deposition of the radiocontaminant per square meter of ground is considered small. Study of the factors involved in transfer of radioiodine from forage to milk is therefore important.

Two experimental approaches have been used to study the secretion of I^{131} in milk. One is the use of single doses of radioiodine, and the other is administration daily for relatively long periods of time. Each approach is of direct value, since a single dose gives information pertinent to a short exposure to radioiodine, such as was experienced at Windscale, and the long-term administration is applicable to situations where nuclear testing or contamination takes place over a considerable period. In addition, it has been demonstrated by double tracer studies using I^{125} and I^{131} that the data of single administration experiments are readily interpretable in terms of long-term administration.(33).

Dairy cattle have been found to secrete in milk from 4 to 20% of a single dose of I^{131} in a 7-day period, the average value being about 8% (35,35,36,37,38,39). After a period of daily administration of I^{131} the total collection of radioiodine in milk of another group of cows was found to plateau at from 4 to 20%, and to average about 11% of the daily intake (38,39). The concentration of I^{131} in the milk ranged from 0.5 to 2.7% of the daily intake per liter, and averaged 1.1% per liter of milk (36,38,39). The experimental determinations have been found to give the same results as forage contaminated with I^{131} (35).

The goat has been investigated as well, since it contributes tothe milk supply in some areas. Single administration studies have shown that the total amount of I^{131} in the milk of the goat can vary from 6 to 54% when collected for a period of at least 7 days (39,40). The variation is due to the extremes in the levels of milk production of the animals used. A reasonable, typical value would be about 40% of the dose of radioiodine appearing in the milk. Again, allowance should be made for the lesser I^{131} intake by the goat, when contrasting the value for goats to that of cows.

In studies employing long-term dosing with I^{131}, a tremendous range, from 1 to 80% of a daily dose, was found in the milk of a goat when equilibrium was attained (39). Again, the average value was 40% of a daily dose appearing in the milk produced daily by goats lactating at a reasonable level. The concentration of I^{131} at

equilibrium ranged from 22 to 150, with an average of 65% of a daily intake of I^{131} per liter of milk.

Little of the I^{131} in the milk of cows and goats is associated with the protein, over 90% being in the form of iodide (38,39,30). Of the products from milk, cream and cream-products should contain little radioiodine, and only about 10 to 15 percent of the radioiodine of the milk should be found in cheese made from whole milk (39).

The cow, when compared to the human being, dog, rat or guinea-pig, shows little ability to concentrate iodine from the blood plasma into milk. Milk-plasma ratios in the cow range from about 1 to 4; the other species show abilities to concentrate iodine by a factor of 20 to 30. The ability of the cow to concentrate iodine in its milk shows a seasonal variation (36,37). This changing ability to concentrate iodine makes it difficult to predict with assurance the total I^{131} or the concentration of I^{131} to be expected in the milk of a cow. The goat also shows an ability to concentrate iodine from plasma in producing milk: the milk-plasma ratio is approximately 10 in this species (39,41). The concentrating mechanism is susceptible to attenuation by drugs, such as thiocyanate, perchlorate, or iodine (35,39,41) and offers possibilities of reduction of the transfer of I^{131} into milk.

More work needs to be done to substantiate the effect of season on the iodine concentrating mechanism, to ascertain the factors which is the ultimate cause of the variation, and to investigate the effects of drugs and hormones so that the mechanism can be understood and manipulation attempted in times of need.

In addition to milk, eggs could under some circumstances be vectors of radioiodine. Experimental studies with laying hens (42) given radioiodine daily for a period of 20 days, showed that the body of the hen tends to reach a plateau level representing about 70% of the daily dose, with about 90% of each day's dose being found in the excretion. About 8% of the daily intake is recovered in the egg, with the major portion of the egg I^{131} being found in the yolk. Most of the I^{131} is extractable with water, but a significant amount is bound to the protein. When the laying hen is given a single dose of I^{131}, the albumen and the shell show a peak of activity on the first day after administration, and decrease steadily after that. The yolk, however, does not reach its maximum value until about the sixth day, indicating that the radioiodine is incorporated into egg yolk, while the follicle is developing

on the ovary. Practically, this implies that the maximum level in eggs will not arise until sometime after the hens have been exposed. It should be apparent that only hens allowed access to range could develop significant levels of radioiodine in their eggs, a situation that is unusual in commercial production areas of the United States.

VI. Turnover of Cesium

The retention pattern of cesium in mammals has been given, as for other radionuclides, in terms of a multi-component exponential equation in the form:

$$R_t = a_1 e^{-b_1 t} + a_2 e^{-b_2 t} \ldots\ldots a_n e^{-b_n t} \qquad (i)$$

where R_t = retention at time t; a_1, a_2, ... a_n are concentration coefficients; b_1, b_2, ...b_n are rate constants of elimination or transfer. Another term that has also been used to indicate the relative rate of removal is the biological half-time (BT 1/2) defined by Richmond (43) as "the time necessary for an animal to lose the first half of its initial body burden by all routes of elimination, regardless of method of administration". If elimination of Cs* were by a single exponential process, BT 1/2 would have a significant meaning and would be equal to 0.693/b of the first term of equation (i). However, experiment has shown that radiocesium is removed by two or more processes, with each having a distinctive rate of transfer and compartment size. Thus, the use of BT 1/2 should be used advisedly and with knowledge of its short-comings; this expression is heavily weighted by the early rapid process of removal which may represent a relatively small compartment size. The more precise expression is given in equation (i), which, unfortunately, contains several terms and numbers that are not as easily handled as the less precise BT 1/2.

Several laboratories have reported the turnover rates and/or biological half-times for radiocesium in a number of species. The most extensive studies were carried on by Richmond (43) at the Los Alamos Scientific Laboratory. Some of these values have

been compiled for this report and are presented in Table 7; where possible, the terms of the multi-component curve as well as the BT 1/2 are given. In certain cases, only the BT 1/2 or a single term of the exponential equation was available. It may be seen that there appears to be a direct correlation between BT 1/2 and the body size of the various monogastric species; i.e. mouse to man. This relationship was quantitated to a first approximation by Richmond (43) who found that:

$$\log y = 1.864 + 0.825 \log x \qquad \text{(ii)}$$

where y = BT 1/2 in days for Cs*, and x = body surface area of the animal in square meters. Although Richmond (43) had not suggested any physiological basis for this interspecies correlation, it was tempting to hypothesize that Cs* turnover may be related to the metabolic body size and, therefore, to metabolic rate of the different species. In preliminary studies in this laboratory, this hypothesis was tested in an experiment in which the effect of thiouracil feeding on Cs^{137} retention in rats was determined (44). Although thiouracil inhibited growth and decreased oxygen consumption in the treated group, the rates of elimination of Cs^{137} by the controls and the thiouracil-fed animals were essentially the same. Thus, the physiological explanation for this relationship must be sought elsewhere.

From Table 7 it was also apparent that the BT 1/2 for the goat and cow was relatively short compared to the non-ruminant species. The longest component for the goat had a half-time of about 3.6 days, whereas man, comparable in body size, had a long half-time of about 144 days. The BT 1/2 for the cow was 4-8 days, with the longest component having a half-time of about 37 days. The reason for these significant differences between animal classes is unknown, but may be related to the normally large intake of potassium by the ruminant, and their physiological mechanisms for handling this alkali metal ion.

The earlier observations of Richmond (43) indicated a BT 1/2 for man of about 110 days; more recent values given by Rosoff *et al.* (45) suggested a much shorter BT 1/2 of 50-60 days. The discrepancy between these values may be due to the type of experimental subject: Richmond (43) had used healthy male laboratory personnel, whereas the subjects of Rosoff's study were older hospital patients.

Attention is called to the possibility that small amounts of radiocesium may be trapped in cartilage or bone and thus not be

TABLE 7

Turnover of Radiocesium (Cs^{134} or Cs^{137}) in Mammals

Species	Method of Administration*	BT 1/2 (days)	A_1	$\frac{0.693}{b_1}$ (days)	A_2	$\frac{0.693}{b_2}$ (days)	A_3	$\frac{0.693}{b_3}$ (days)	Ref.
Mouse	Intraperitoneal	1.2	32.4	0.5	46.0	2.4	21.6	6.6	(43)
Rat	Oral	6.5	17.0	0.8	41.0	6.8	42.0	13.5	(43)
Rat	Intraperitoneal	---	23.0	1.5	32.0	7.0	45.0	14.0	(43)
Rat	Oral	7.5	7.8	0.7	28.3	2.4	64.9	15.7	(44)
Rat	Intraperitoneal	---	---	---	70	7	30	16	(49)
Monkey	Intravenous	19.0	18.0	3.0	60.0	22.3	22.0	40.8	(43)
Dog	Intravenous	25.0	14.0	1.1	56.0	27.0	30.0	43.6	(43)
Dog	Oral	---	---	---	77	18.8	---	---	(50)
Dog	Oral,	---	---	---	85	40.9	---	---	(50)
Dog	Oral, multiple	---	---	---	67-75	22-30	---	---	(50)
Man	Oral	110.0	16.0	3.0	84.0	144.3	---	---	(43)
Man	Intravenous	50-60	---	---	---	---	---	---	(45)
Goat	Intravenous	---	54	0.7	46.0	3.6	---	---	(51)
Cow	Intravenous	8.0	24.0	2.1	28.5	4.9	47.5	37.3	(46)
Cow	Oral	4.0	---	---	---	---	---	---	(46)

* Single dose unless otherwise specified.

available for exponential removal (46,47,48).

In regard to the comparative behavior of cesium and potassium, further evidence on man (48a) and on rats (48b) supports the lack of interdependence of these two elements.

VII. Transfer of Radiocesium into Milk

As for strontium, milk represents a significant pathway for transport of cesium to the human population. Laboratory studies have shown that from 8 to 13% of a single ingested dose will be recovered in the milk of a dairy cow during a subsequent collection period of about 7 days (46,52,53). These data emphasize that radiocesium is more readily secreted into milk, by a factor of at least 10, than is radiostrontium.

In simulating conditions of a stratospheric reservoir of fallout, experiments in which radiocesium was given to dairy cattle every day for 30 days showed that a plateau in milk cesium was not quite attained during this period, and that from 11 to 15 % of the daily intake could be recovered in the daily production of milk (54). From 1.3 to 1.5% of the daily intake was found per liter of milk at this time. The data from the single dose experiments agree with this latter value when the single dose data are calculated on the basis that the summation of the recoveries of radiocesium in milk on at least 7 successive days following a single dose will be equal to the plateau value observed after a long period of daily administration of the radioisotopes.

The feeding of about 10 grams of stable cesium per day to the cow had no effect upon the amount of ingested radiocesium that was secreted in the milk.

For the goat, about 9.4% of its daily intake of radiocesium was found in each liter of milk (51). The lower intake of radiocesium by the animal must be accounted for before any special significance is put upon this figure.

Since cesium is chemically similar to potassium, some attempts have been made to correlate the behavior of the two elements. The cesium/potassium ratio from diet to milk has been shown to be 1.6 for dairy cows and 1.3 for lactating goats (51,52). This preferential secretion of cesium does not appear to be due to the mammary gland since the plasma ratios were the same as that of

the milk (51).

More work is needed in regard to the passage of cesium into milk. One particular approach should be to estimate the relative constancy of the percent of the daily intake of radiocesium that is secreted per liter of milk. If the value is sufficiently constant it may be possible to determine with some accuracy the amount of cesium being consumed daily by dairy cows. With some knowledge of feeding practices it may be possible to correlate milk cesium with the rate of return of fission products from the stratosphere.

Other problems that are of practical importance include: (a) methods to reduce the amount of cesium secreted into milk, (b) methods of removal of cesium after incorporation into milk, and (c) the retention of cesium when butter, cheese, cream, skim-milk and other processed products are made from the milk.

Summary

1. This paper presents a status review on selected aspects of food surveys, strontium, cesium and iodine as related to fallout problems.

2. Knowledge for dealing with the "fallout problem" must come from investigations at all steps of a process that can be represented as follows:

   (a) release of radiocontamination
   (b) distribution in soil, air, water
   (c) amounts of radioactivity in different foods
   (d) amounts of radioactivity in total diet
   (e) amounts and distribution of radioactivity in human population
   (f) radiation doses to population
   (g) harm to population

3. The design and scope of food surveys should be based on the required objectives. The representativeness of sampling and precision of assay required is governed by official acceptable levels. An important waste of effort and expenditure may result from attempts to provide accurate evaluation of levels that are small compared to those of significance.

   Survey results to-date lead to the suggestion that rigorous

sampling procedures may not be justified for many practical purposes.

4. $OR_{body/diet}\left(\frac{\text{Sr/Ca of body}}{\text{Sr/Ca of diet}}\right)$ values for adult man range from 0.16 to 0.5, with values based on most extensive sampling falling near 0.25. Values are probably higher, 0.5 and above, for the very young; the significance of the higher values in respect to harm cannot yet be assessed.

5. $OR_{milk/diet}$ values for man, cow and goat are about 0.1. On typical rations the concentration of fallout Sr^{90} in milk was halved by doubling the calcium content of the diet in a long-term study. This confirms predictions from short-term experimental studies.

6. Secretion of radioiodine into milk is highly variable. Typically the dairy cow secreted about 1% of the ingested daily intake per liter of milk. Attention is called to the differing abilities of various species to concentrate iodine from blood plasma to milk.

   About 8% of the daily radioiodine intake is found in the egg of the laying hen, with most being found in the yolk and a significant porportion bound to protein. After a single administration the yolk does not reach a maximum until about the sixth day.

7. The turnover time of cesium in various species is reviewed, and the need stressed for consideration in terms of multi-component exponential behavior. In monogastric species the overall turnover time appears to correlate with body size, ranging from a few days for the mouse to about 100 days for man. In ruminants the values are relatively smaller, being a few days for the goat and somewhat longer for the cow.

   Special note is made of the possibility that small amounts of radiocesium may be trapped in the skeleton and thus not be available for exponential removal.

In the dairy cow, about 1.5% of the daily ingested radiocesium was found per liter of milk. Thus, ingested radiocesium is secreted about ten times more effectively into milk than is radiostrontium. Cesium is secreted into milk about 1.5 times more effectively than ingested potassium.

Feeding of 10 grams of stable cesium per day to the cow had no effect on the amount of radiocesium secreted into milk.

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PREDICTION OF THE SR^{90} CONCENTRATION IN THE WORLD POPULATION

J. Laurence Kulp
Lamont Geological Observatory
Columbia University
Palisades, N. Y.

Introduction

The ultimate goal in fallout research is prediction of the concentration of the radioactive nuclides produced in nuclear detonations in the human environment. To achieve this goal the mechanism of dissemination of the radioactive debris throughout the world and into the human body must be defined. Although the accuracy of the predictive apparatus can be continually improved by further investigations, the current state of knowledge appears to be sufficient so that the errors in the prediction are less than the uncertainties in the biological effects. The errors in prediction also appear to be small compared with the possible modification of these concentrations by modern technology available to society.

The basic parameters involved in the prediction of the amount of fallout or the concentration in the human body of any nuclide at any time and place are:

(1) the quantity and nature of the debris introduced into the atmosphere, (2) the time, location and altitude of the injection, (3) the pattern of dispersion of the debris throughout the atmosphere and its rate of descent to the lower atmosphere, (4) the mechanism of removal of the debris from the lower atmosphere to the ground, (5) the nature of the uptake of the fallout nuclides into foodstuffs, (6) the efficiency with which these nuclides are deposited in human tissue, and (7) the residence time of the nuclides in the body.

This report (1) is concerned only with the problem of estimating the concentration of Sr^{90} in the world population at some future time after a given input of debris. Many of the principles hold for most of the other nuclides as well.

Major research programs have been directed toward the definition of these parameters. The High Altitude Sampling Project of the Department of Defense (2) supplemented by the balloon data on Project Ashcan (3) has provided the information on the stratospheric dispersion and removal. The pattern of deposition on the surface of the earth and the rates of removal have been defined by the rainfall network of the AEC Health and Safety Laboratory (3) and the soil program of the U. S. Department of Agriculture (3). The movement of the Sr^{90} into the food chain and its concentration in diet has been studied by a number of laboratories (4, 5, 6). The uptake in the human skeleton as a function of diet, geographical location, time, and age of the individual has been studied in this laboratory with the aid of a world-wide network of bone sampling stations, which over an 8 year period have supplied over 10,000 samples of human bone for analysis.

Deposition from Nuclear Explosions in the Atmosphere

For the purpose of estimating future Sr^{90} levels in the human population as a result of given nuclear explosions, it is necessary to know the fraction of the debris that enters the stratosphere, the time it stays there and the pattern of surface deposition. Only MT explosions will be considered. The Mc of Sr^{90} produced is about 1/10 of the fission yield in MT.

The clouds from all MT explosions in the atmosphere will stabilize in the stratosphere. Those exploded at sufficient altitude such that the firefall does not touch the ground will inject essentially all of their radioactive debris into the stratosphere. If the explosion occurs at the land surface, a major fraction of the debris will fall out locally. The fraction introduced into the stratosphere that will contribute to world-wide fallout is estimated to be about 20% although the uncertainty on this number is large (2).

The fate of the debris once it has entered the stratosphere has been studied by the High Altitude Sampling Project (2). The rate of mixing and removal depends largely on the location of the stabilized cloud. In general, it has been found that horizontal mixing is much more rapid than vertical mixing, that clouds from the same yield explosions will stabilize at lower altitudes in the polar stratosphere than in the lower atmosphere, that vertical mixing is much more rapid in the polar stratosphere than the tropical stratosphere to the troposphere is by horizontal turbulent mixing and organized meandering of air currents through the tropopause gap region.

Although the movement of debris in the stratosphere is a complex matter, only the gross relationships are of concern in predicting cumulative fallout levels. The concept of half-residence time while not strictly correct since it is not constant for even a given cloud, is quite useful since the approximate time required for the first half of the debris introduced at almost any point can be estimated from existing data. This first half

defines the peak concentration in the rate of fallout after a nuclear event in the atmosphere and carries most of the effect of the shorter lived isotopes. The second half of what remains, bringing the total deposition to 75% may be 1.5 times the first "half-residence time" but this is not crucial in the overall prediction. Even if the rate of transfer were to decay exponentially the significant deposition would have occurred in two or three "half-times".

Table 1 summarizes the "half-residence times" for the first half of any cloud to be transferred to the troposphere as a function of the location at stabilization. Also given is the degree of asymmetry to be expected in the Northern and Southern Hemispheres. It is seen that the range in time is from 3-5 months in the lower polar stratosphere to as much as 5 years for explosions in the mesosphere. Since all of these times are short compared with the radioactive half-life of Sr^{90}, it means that essentially all of the debris will be deposited and enter the biological sphere. Clouds that stabilize in the lower stratosphere will be deposited fast enough so that the rate of fallout may be important. With regard to latitude, explosions at the equator should deposit symmetrically regardless of the altitude of cloud stabilization. The other extreme is

TABLE 1

Half-residence times for various injections into the stratosphere and associated hemispheric asymmetry of deposition

Latitude	Altitude (ft.)	Half-Residence Time (first half of debris)	Ultimately % Deposited in Southern Hemisphere
60-90°N	Lower Stratosphere 30,000-80,000	3-5 mos.	<10
	High Stratosphere 80,000-150,000	1-2 yrs.	<25
12°N	Lower Stratosphere 60,000-80,000	8-10 mos.	~33
0°N	Lower Stratosphere 60,000-80,000	8-10 mos.	50
	High Stratosphere 80,000-120,000	~3 yrs.	50
	Mesosphere 150,000+	~5 yrs.	50

represented by clouds introduced into the lower poler stratosphere. In this case it appears that less than 10%, and quite possibly less than 5%, of the debris will enter the Southern Hemisphere. With increasing altitude, world-wide mixing increases. Clouds introduced into the lower stratosphere 30-60°N should deposit at least 80-85% of their debris into the Northern Hemisphere. The factors of uncertainty in either time or place for the bulk of the debris are not large.

The tropopause gap through which most of the stratospheric debris is introduced into the troposphere occur roughly 30-60°N. Once in the troposphere, the debris is washed out by rain. The amount of deposition being approximately proportional to rainfall (7). Some small fraction may be deposited as dry fallout but this appears to be less than 10% in the areas of 40 in. rainfall. An analysis of the concentration of radioactivity in rain as a function of latitude and the deposition of the pre-1953 Nevada (tropospheric) debris leads to the conclusion that south of 30°N the fallout decreases by a factor of two with each 10° latitude band to the equator. This is a result of the competition between the lateral mixing rate of the tropospheric air and the rate of removal of the debris by precipitation. Both of these rates appear to have half-times of 3 to 4 weeks. North of 60°N the average mean annual rainfall drops off rapidly with the result that the concentration in rain and air remain similar to that in the 30-60°N belt.

Based on these studies it is possible to estimate the Sr^{90} deposition in $mc/mi.^2/in.$ of rainfall for each 10° latitude zone for each Mc of Sr^{90} that will be deposited in that hemisphere. These results are given in Table 2. The combination of the data of Tables 1 and 2 permit an estimate of the rate of fallout and the cumulative deposition to be expected at any specified location if the mean annual rainfall is known. The smaller the area chosen, the larger the uncertainty; but if say, the deposition of all areas 30-40°N that have 40-60 in. of mean annual rainfall are specified, the errors should be less than 30%.

TABLE 2

Approximate Sr^{90} deposition with latitude

Latitude Band	mc $Sr^{90}/mi.^2/in.$ of mean annual rainfall*
30-90°N	0.25
20-30°N	0.12
10-20°N	0.06
0-10°N	0.03

*For 1 Mc of Sr^{90} transferred into the northern troposphere.

Relation of Deposition to Sr^{90} Content of Milk

Sr^{90} enters the plants which the cow uses for food via two routes: direct absorption and uptake through the soil. The direct absorption is proportional to the rate of fallout during the growth of the plants while the uptake through the soil is proportional to the cumulative deposit on the soil, assuming a fixed vertical profile, soil type and plant. Several years after a nuclear explosion in the atmosphere the Sr^{90} in milk and the rest of the diet is supported only by cumulative deposit. In the first year, however, the direct absorption may be the dominant source of Sr^{90}, unless the cumulative deposit is very large compared to the fallout. The importance of direct absorpiton was first cogently set forth by Russell (8).

The number of lines of evidence supported the observation that direct absorption played an important role, but at the time of our last report (9), the only quantitative data were those of Burton, Milbourn and Russell (10), on certain experimental fields in the United Kingdom. These data indicated that in 1958 the rate factor accounted for about 80% of the Sr^{90} in the milk produced in England and Wales. In more recent studies, these workers (11) revised their estimate of the contribution of the rate factor in 1958-59 to 60-70%, and in 1960 estimated the contribution to be only 30%.

It was pointed out in the last report (9) that, in order to obtain a reasonably accurate definition of the relative importance of the rate and cumulative fallout factors for the North American population, it would be necessary to observe the milk levels throughout North America for 1959 through 1960, when the rate of fallout was expected to drop by about a factor of 5 while the cumulative deposit was expected to remain nearly constant. The data for such a comparison are now available (Table 3).

For a given field, the relationship between the cumulative deposit of Sr^{90}, the rate of fallout during the growing period of the grass, and the ratio of Sr^{90}/Ca in the milk that is produced is a very complex one. The ultimate concentration in the milk depends on the physical and chemical characteristics of the soil, the type of grass, the stage of growth, the precipitation pattern during the period of growth, the concentration of Sr^{90} in each rain, and the cumulative deposit of Sr^{90}. The average for each of these factors, however, should be reasonably constant over large areas, such as the eastern U. S., and for sufficiently long periods of time. For such a situation the following relationship should hold:

$$\underline{Q_M} = \underline{AX} + \underline{BY}$$

TABLE 3

Data for calculating the contribution of the rate of fallout and the cumulative deposit to Sr^{90} levels in milk in the U. S. and Canada

Period	Sr^{90} concentrations		
	Milk* ($\mu\mu c\ Sr^{90}/g\ Ca$)	Rain** ($mc/mi.^2$)	Earth's surface*** ($mc/mi.^2$)
	Mean annual rainfall, 40-60 in.		
Apr.-Oct. 1957	6.2	6.4	23
Apr.-Oct. 1958	11.4	8.1	38
Apr.-Oct. 1959	15.6	11.3	61
Apr.-Oct. 1960	9.7	2.5	65
Apr.-Oct. 1961	8.7	2.4	69
	Mean annual rainfall, 30-40 in.		
Apr.-Oct. 1957	6.7	4.3	22
Apr.-Oct. 1958	9.6	10.3	37
Apr.-Oct. 1959	13.3	10.3	59
Apr.-Oct. 1960	8.8	2.3	62
Apr.-Oct. 1961	6.9	2.1	65

* Averages for the 6-month growing season 1 April to 1 October (see Table 5).

** Totals for the 6-month growing season 1 April to 1 October.

***Averages for cumulative deposit on 1 July, the mid-point of the growing season.

where Q_M is the average number of μμc Sr^{90}/g Ca in milk for a given large area of fairly uniform mean annual rainfall and average concentration in rain during the growing season; X is the average rate of deposition of Sr^{90} (in mc/mi.^{2}) per 6 months for a particular growing season; Y is the cumulative deposit of Sr^{90} (in mc/mi.^{2}) measured at midpoint of the growing season; A is the number of μμc Sr^{90}/g/Ca in milk, due to direct absorption, divided by the number of mc Sr^{90}/mi.^{2} deposited during the growing season; and B is the number of μμc Sr^{90}/g Ca, due to the cumulative deposit, divided by the total mc Sr^{90}/mi.^{2} deposited to the midpoint of the growing season since the start of nuclear testing.

In order to compute the constants A and B, data from two rainfall zones in North America which contain the largest number of milk, soil and rainfall stations were used. The zone of 40 to 60 in. of mean annual rainfall includes most of the eastern U. S. and is roughly bounded on the west and north by the Mississippi, Ohio and St. Lawrence Rivers. It also includes a smaller area along the coast in the Pacific Northwest. The zone of 30 to 40 in. of mean annual rainfall is roughly bounded on the west and north by a line passing through Houston, Tulsa, Kansas City, Duluth and Goose Bay, Labrador. Throughout these large areas, it is assumed, the effective growing season is the 6-month period from 1 April to 1 October. The data are assembled in Table 3. The levels in milk are taken from Table 4 which compiles all available analyses (12). The cumulative deposition for 1 July of each year is taken from Alexander's collection (3) for 1957, 1958, and 1959, with extrapolation to 1960 on the basis of known rainout. The average deposition each year from 1 April to 1 October was obtained by multiplying the average concentration in rain for the latitude band 30° to 70° N (2, 3) by the average rainfall for the 6-month period.

The most accurate estimates of A and B can be made by solving the foregoing equation for 1959 and 1960, since the most drastic change in X and Y occurred in these two years. The results for the 40- to 60- in. zone are A = 0.73 + 0.10 and B = 0.12 + 0.01, results for the 30- to 40- in. zone are A = 0.58 + 0.08, B = 0.12 + 0.01. The constants A = 0.65 and B = 0.12 were then adopted, and the levels in milk for each year from 1957 through 1961 were calculated. The results are given in Table 5 and compared with the observed levels in milk. The error with respect to the observed levels is 5 to 10%. The error with respect to the predicted levels is 10 to 20%. It may be seen that calculations on the basis of these constants predict the levels in milk for 1957, 1958 and 1961 within the experimental uncertainities. The accuracy is limited largely by the error in the deposition rate. The number of rain stations is much smaller than the number of milk or soil stations in 1958-60. The data for 1957 are scarce.

The percentage of the Sr^{90} in milk of these areas that is due to the rate of fallout (direct absorption) is also shown in Table 5. Thus, during the period 1957-59, with successive increases in the rate of

TABLE 4

Concentrations of Sr^{90} in milk from North American stations in areas of different average annual precipitation (figures in parentheses are number of stations)

Quarter	Concentration ($\mu\mu c$ Sr^{90}/g Ca)				
	40-60 in.	30-40 in.	20-30 in.	10-20 in.	0 to 10 in.
		1957			
1	4.6 (4)	5.1 (10)	3.9 (6)	3.5 (9)	1.4 (3)
2	7.7 (7)	6.2 (16)	7.0 (11)	5.5 (12)	2.3 (4)
3	7.5 (7)	7.6 (18)	6.7 (11)	8.6 (11)	1.8 (4)
4	8.8 (7)	6.8 (16)	9.3 (13)	8.2 (12)	1.7 (3)
Calendar year av.	7.5	6.6	7.2	6.5	1.8
Av. for 1 Apr. 1957 to 31 Mar. 1958	8.2	6.9	7.5	7.5	2.1
		1958			
1	8.8 (7)	6.9 (16)	7.2 (15)	7.8 (12)	2.2 (4)
2	12.3 (8)	9.2 (19)	9.2 (18)	9.3 (12)	3.4 (4)
3	10.4 (19)	10.0 (26)	9.5 (24)	8.3 (16)	4.1 (8)
4	10.8 (10)	10.7 (22)	10.0 (22)	8.4 (12)	2.0 (3)
Calendar year av.	10.6	10.0	9.1	8.5	3.2
Av. for 1 Apr. 1958 to 31 Mar. 1959	11.7	10.1	9.7	8.6	3.7

(Continued)

TABLE 4 (Cont'd)

Quarter	Concentration ($\mu\mu$c Sr^{90}/g Ca)				
	40-60 in.	30-40 in.	20-30 in.	10-20 in.	0 to 10 in.
		1959			
1	13.7 (9)	10.6 (16)	10.4 (17)	8.5 (11)	4.6 (3)
2	19.0 (9)	15.8 (18)	14.6 (18)	12.2 (12)	8.0 (3)
3	12.2 (17)	10.8 (23)	10.2 (18)	9.8 (16)	2.9 (4)
4	11.5 (11)	10.1 (17)	9.9 (15)	10.1 (14)	2.6 (5)
Calendar year av.	13.6	11.3	11.3	8.5	4.1
Av. for 1 Apr. 1959 to 31 Mar. 1960	13.5	12.0	11.5	10.3	3.4
		1960			
1	12.5 (6)	11.2 (15)	10.9 (19)	9.7 (15)	2.3 (7)
2	10.6 (12)	10.1 (25)	10.2 (14)	8.9 (15)	3.8 (4)
3	8.7 (20)	7.5 (29)	7.4 (16)	6.2 (14)	3.1 (7)
4	7.8 (19)	6.8 (24)	7.5 (13)	6.5 (12)	3.2 (4)
Calendar year av.	9.2	8.6	7.9	7.9	3.0
Av. for 1 Apr. 1960 to 31 Mar. 1961	8.3	7.7	7.3	6.9	3.1
		1961			
1	6.9 (19)	5.9 (21)	4.0 (5)	4.5 (6)	2.2 (4)

as well as the cumulative deposit, the contributions from the rate and cumulative-deposit factors were roughly equal. Our earlier suggestion (9) that the rate factor was dominant during 1954-55 is therefore incorrect. Direct absorption may have been the dominant mechanism of the entry of Sr^{90} into the food chain only in 1954-55. In 1960 the contribution of the rate factor (direct absorption) dropped to about 15% of the total. In 1961 the cumulative deposit increased about 5% and the rate of fallout was similar to that in 1960; hence the level of Sr^{90} in milk should have increased by less than 5%. This would lie within present experimental and sampling errors.

If the average levels in milk and the average rates of deposition for the entire year instead of the 6-month growing period are used in the calculation, the results are not qualitatively different. By limiting the comparison to the growing period, the factors may be more closely controlled. During the fourth quarter of one year and the first quarter of the next year, the cows largely consume fodder produced in the preceding growing season. Thus, the levels in milk remain fairly constant for these quarters, whereas the fallout rate may change. The effect of undiminished or increased rates of fallout during the winter would primarily serve to increase the cumulative deposit for the next growing season.

Knapp (13) has also calculated the rate and cumulative coefficients. His method appears less reliable than that used above for three reasons: (i) only a fraction of the available milk data were used, i. e. those collected by the U. S. Public Health Service network (14), (ii) the milk, rainfall, and (iii) the coefficients were computed by comparing two arbitrarily chosen months, one in 1959 and one in 1960. The concentration of Sr^{90} in milk in highly variable over short time intervals or small goegraphical areas. To obtain the most meaningful average values for the coefficients it appears necessary to compare whole growing seasons over large section of a continent that have similar rainfall and, therefore, similar cumulative deposits and rates of fallout.

Relation of Milk to Total Diet

The concentration of Sr^{90} in the total diet is best approximated in a population of Western culture by multiplying the predicted milk level by a constant factor which has been determined experimentally for a number of regions.

The ratio of Sr^{90}/Ca for the total diet to that for milk in the Western diet has been studied initially by Kulp and Slakter (4) for the U. S. by Bryant et al. (5) for the United Kingdom, and by Bird and Mar (15) for Canada; these workers found a value in 1957-58 of about 1.2. Subsequent studies by the AEC Health and Safety Laboratory, Consumers Union, and the U. S. Public Health Service (16) gave values ranging from 1.21 to 1.32 for various U. S. cities. The most comprehensive study, by

TABLE 5

Contribution of the rate of fallout and the cumulative deposit to Sr^{90} concentration in milk in the U. S. and Canada (see text)

Year	Sr^{90} Concentrations in Milk (μμc/g Ca)				
	AX	BY	Total	Observed	% Rate
		Mean annual rainfall, 40-60 in.			
1957	4.2	2.8	7	6	60
1958	5.3	4.6	10	11	53
1959	7.4	7.3	15	16	50
1960	1.6	7.8	9	10	17
1961	1.6	8.3	10	9	16
		Mean annual rainfall, 30-40 in.			
1957	2.8	2.6	5	7	52
1958	6.7	4.4	11	10	60
1959	6.7	7.1	14	13	49
1960	1.5	7.4	9	9	17
1961	1.4	7.8	9	7	15

Consumers Union on eight cities, gave 1.21. In the United Kingdom the ratio of Sr^{90}/Ca for the diet to the ratio for milk appears to be closer to 1.0 as a result of the addition of mineral Ca to the national diet (11). It is possible that the ratio of Sr^{90} in total diet to that in milk will vary as the relative contributions of direct absorption and soil uptake change but by the summer of 1961 when these contributions had been greatly altered there was no evidence in New York or Chicago of any major change (17).

Discrimination Factor from Diet to Bone

The discrimination factor against Sr in favor of Ca is about 4 for adults on average Western diets (5, 6, 18). This factor is probably independent of age except possibly in the case of children under 1 year. Bryant and Loutit (19) suggest that in the first year of life the discrimination factor may be as low as 2.5 to 2.0. Their suggestion is based on two arguments: (i) that determinations of common Sr in diet and bone show a discrimination factor of about 2.5, and (ii) that the best fit of their 1959 bone data requires a discrimination factor of less than 4 for the first 2 years. On examination neither argument appears conclusive. The determinations of common strontium in the diet were made in 1939. The sources of the foodstuffs in the diet in 1939 may have been very different from the sources of foodstuffs which produced the bones which were analyzed. Moreover, the bone samples used by Bryant and Loutit were largely from Wales where the diet levels are much higher than in the eastern U. S. on which the comparative curve was calculated. If the average Welsh diet were used, the discrimination factor of 4 appears more appropriate.

The Bone Curve

It has been shown in earlier work (9) that a relatively simple model for the uptake of Sr^{90} by the bones of young people can account for the observed distribtion with age. This treatment was suggested originally by Langham and Anderson (20).

The present model is based on two assumptions, (i) that the rate of exchange of existing bone is independent of age, and (ii) that the discrimination factor between Sr and Ca in passing from diet to bone is independent of age. In order to calculate the Sr^{90} concentration in the bones of any age group that subsist on the same average diet, the following experimental data are required: (i) the concentration of Sr^{90} in the fetus and the adult, (ii) the Sr^{90} concentration in the diet for each year, (iii) the amount of Ca added to the skeleton per year at each age, and (iv) the discrimination factor between Sr and Ca, i.e. $\left(\frac{Sr}{Ca}\right)$ Diet / $\left(\frac{Sr}{Ca}\right)$ Bone. The equation for the concentration of Sr^{90} in individuals of each age then is:

$$Q_B = \frac{\sum_{i=1}^{p} Ca_{n_i} A_{n_i} + Ca_{ex_i} (A_{n_i} - A_{s_{i-1}}) - 0.025\, Ca_{s_{i-1}} A_{s_{i-1}}}{Ca_{s_p}}$$

where Ca_n is the number of g of Ca added in a given 12-month period in the formation of new bone (a value which depends on the age of the individual); Ca_{ex} is the number of g of Ca exchanged per year: Ca_s is the number of g of Ca in the skeleton; Ca_{s_p} is the total number of g of Ca in the present skeleton; A_{n_i} is the specific activity of newly depositing bone, in $\mu\mu c\ Sr^{90}/g$ Ca; A_{s_i-1} is the specific activity of the total skeleton at the mid-point of the given calendar year. The summation is over all years for which there was measurable Sr^{90} in the diet. The factor 0.025 accounts for the radioactive decay of the Sr^{90} fixed in the skeleton the previous year.

Fig. 1 gives the curves calculated for the years 1958 and 1960. The values for fetal samples represent the experimental averages for all samples from North America, but these were predominantly from New York and Chicago. The adult samples were predominantly from New York, Boston, and Houston, with additions from Anchorage in 1960. The rate of exchange was taken as 2.5% as discussed above. The discrimination factor was taken as 4. The amount of Ca added at each age is that obtained by Mitchell *et al.* (21). The concentration of Sr^{90} in the total diet from July to July 1 were (in $\mu\mu c\ Sr^{90}/g$ Ca): 1953-54, 1.5: 1954-55, 3.2; 1955-56, 4.2; 1956-57, 6.3; 1957-58, 10.2; 1958-59, 16.2; 1959-60, 14.1 and 1960-61, 9.6. Some of these factors require further discussion.

The absolute value of the rate of exchange is probably known to ± 20% in adults. In young people the direct addition of Sr^{90} in new bone is much greater than the addition from exchange assuming that the rate of exchange is independent of age. Bryant and Loutit (19) have questioned the constancy of the rate of exchange with age and suggest that the exchange may be 100% during the first year and may drop to nearly zero during puberty. Their argument in support of these two suggestions is inconclusive, due to the relatively large uncertainties in their data. Further, the uniformity of Sr^{90} levels in the different bones of the skeletons of young people (9, 19) suggests that the contribution to the total specific activity from exchange must be small. If the rates of exchange in vertebrae and in the femur shaft differ by a factor of 4 in young people, as they do in adults, and if the average rate of exchange significantly exceeds a few per cent, differences among the bones of one body would be easily observable. Finally, since the discrimination factor from diet to bone is so much greater between the mother's diet and the bone of the fetus than between the diet and the bone of the 1

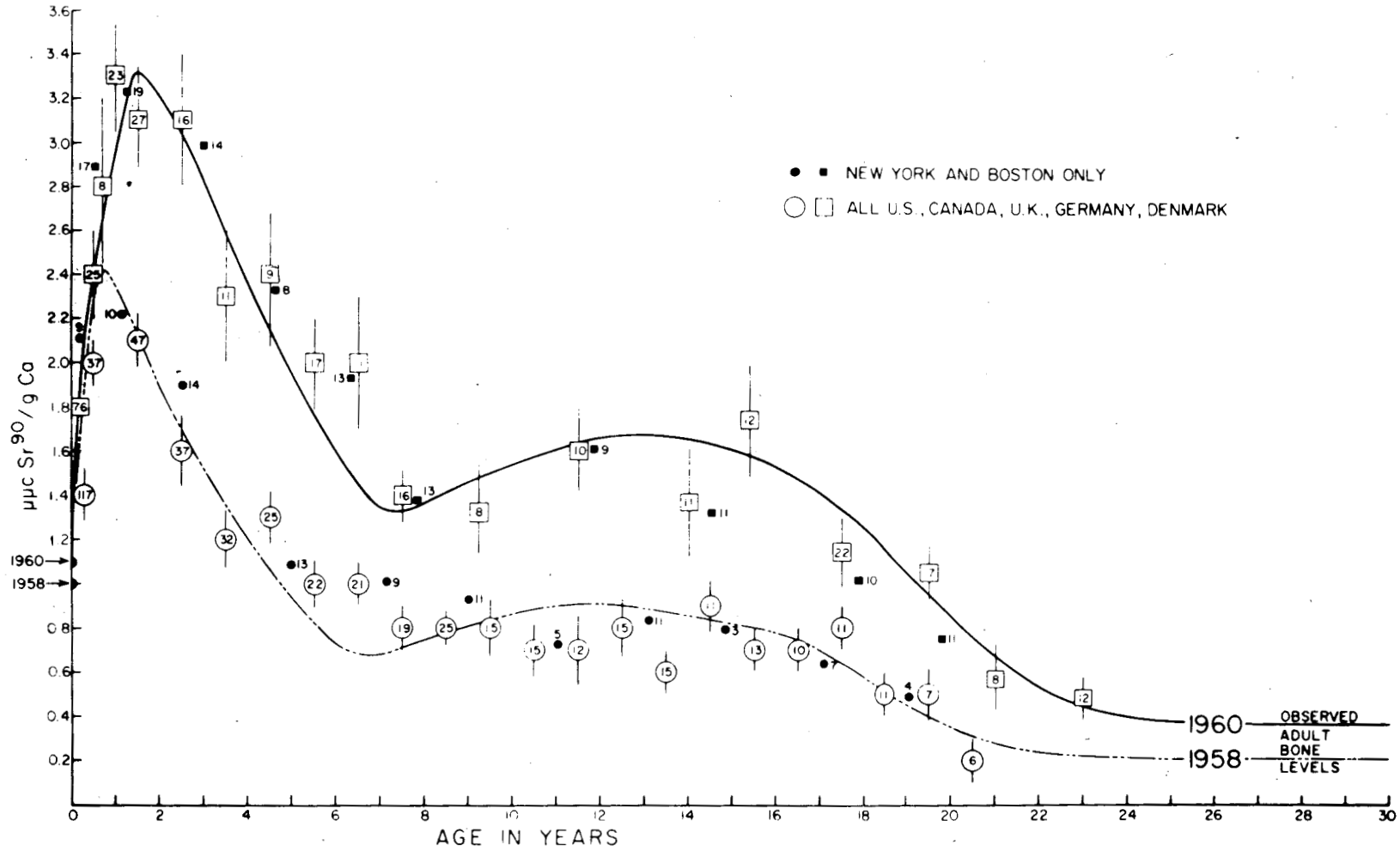

Figure 1. Curves for calculated values for Sr^{90} concentrations in bone in eastern North America, 40-60 in. mean annual rainfall, for 1958 and 1960. The average values of two sets of bone samples (New York and Boston; North America and western Europe) are plotted for comparison with the curves. The numbers associated with the points indicate the number of samples and the length of the vertical line represents the standard error on the mean.

year-old, complete replacement in the first year would yield a much higher specific activity than is actually observed. Therefore, on the basis of present knowledge there seems no reason to adopt a more complex hypothesis concerning the basic physiological process of Ca replacement in the bone.

The value for the Sr^{90} level in diet that was used in these calculations was derived by taking the average level in milk for the eastern U. S. (Table 4) (that is, the zone of 40 to 60 in. of mean annual rainfall) weighted for population, and then multiplying by 1.2. These averages for milk should be representative of the milk consumed by the people of Chicago, New York, and Boston. The data for the period 1953-56 are taken from earlier work (9) which is based on less comprehensive sampling tha th represented by the data in Table 4, but larger uncertainties in the diet levels in the early years of lower specific activity do not significantly affect the computations for the period 1958-61.

In Fig. 1 all of the measurements on samples from North America and western Europe north of latitude 30°N have been plotted for 1958 and 1960. The teen-age samples are corrected for bone type, since it is assumed that in the exchange process the relative rate of turnover of the different bones is the same in teen-agers as in adults. The major stations in North America are New York and Boston. The major stations in the rest of the world are London, Glasgow, and Bonn. In Fig. 1 the solid circles and squares represent the New York and Boston data only. These should give the best fit if the model is correct, since the model was based on the diet in eastern North America. The combined data from Europe and North America appear to fit equally well. The average diets of these sets of individuals must be similar, since the fetus levels agree, falling within about 20% of the mean, the level for Bonn being higher and that for the United Kingdom lower. The same relation appears to hold for the adults. The model appears to fit the data closely for the age period 0 to 4 years.

If the true discrimination factor, $\left(\frac{Sr}{Ca}\right)_{\text{Diet}} / \left(\frac{Sr}{Ca}\right)_{\text{Bone}}$ for the period 0 to 1 year was 2 instead of 4, the observed values for 1-year olds would be close to 6 $\mu\mu c$ Sr^{90}/g Ca, rather than 3.4 (Fig. 1). Furthermore, the peak would not shift to the right on decrease in the levels in diet, as it is observed to do from 1959-60 if the discrimination factor changed rapidly from 2 to 4 during the first 2 years of life. It is concluded that, on present evidence, the simple assumption of a constant discrimination factor from birth to adulthood adequately describes the data. If the discrimination factor is actually less than 4, then another factor, such as systematic variation in the diet, must be operative to compensate.

The data in Fig. 1 fall slightly above the curve in the range 5 to 7 years and slightly below in the range 10 to 15 years. Whether this may merely be due to errors in the assumed additions of Ca to the skeleton at each of these ages or whether it is due to some physiological phenomenon

cannot be stated at this time. In view of the available number of samples, the standard error on the mean (shown by the vertical lines in Fig. 1), and the purpose of the analysis, the correlation is adequate. To study this model in greater detail it would be necessary to obtain about 100 samples in each age from a population whose diet had been adequately sampled over a number of years.

Distribution Within a Population

The above analysis makes it possible to estimate the concentration of Sr^{90} in skeleton in the average person of any age at any locality. It remains to state something about the distribution of the population of individuals of the specified age around this mean.

In Fig. 2 the total population of the New York whole skeleton samples are plotted on probability paper with the assumption of a log-normal distribution. These samples represent a population of nearly 500 individuals. They are all adults so that as has been shown earlier (9) the Sr^{90} concentration does not vary with age. They were all normalized to 1958 prior to plotting in this way. It may be seen that this function fits the data fairly well over the interval from 0.2 to 70% of the population. There is no theoretical justification for this distribution, and other more complex functions could also fit the data over such an interval. Large extrapolations are therefore unwarranted. To use such a curve to predict the distribution beyond 0.001% would appear to be unjusjified.

The curve of Fig. 2 indicates that levels for 5% of the population will exceed twice the average, levels for 0.1% will exceed four times the average, and environment trend to equalize the diet over a long period of time, it seems probably that the foregoing figures represent a conservative estimate of the situation. The distribution curve (Fig. 2) derived experimentally for adults should apply to persons of all other ages since the only significant difference in diet with age would lie in the milk component which shows least variation in its annual average Sr^{90} concentration.

Distribution curves derived from the single-bone analyses are not as meaningful in estimating the actual distribution, since these data have much larger analytical uncertainties and correction must be made to obtain the whole-skeleton burden. The distribution curve for single-bone samples from various population groups that have similar average levels of activity may be useful, however, for purposes of comparison. Such comparisons have been made on the single-bone data from all stations. It is concluded that within the uncertainties of the data, there is no major difference between the distribution curves for urban areas, whether these are areas of Western or of Eastern culture, or in the Northern or Southern Hemisphere. It is thought, however, that in rural

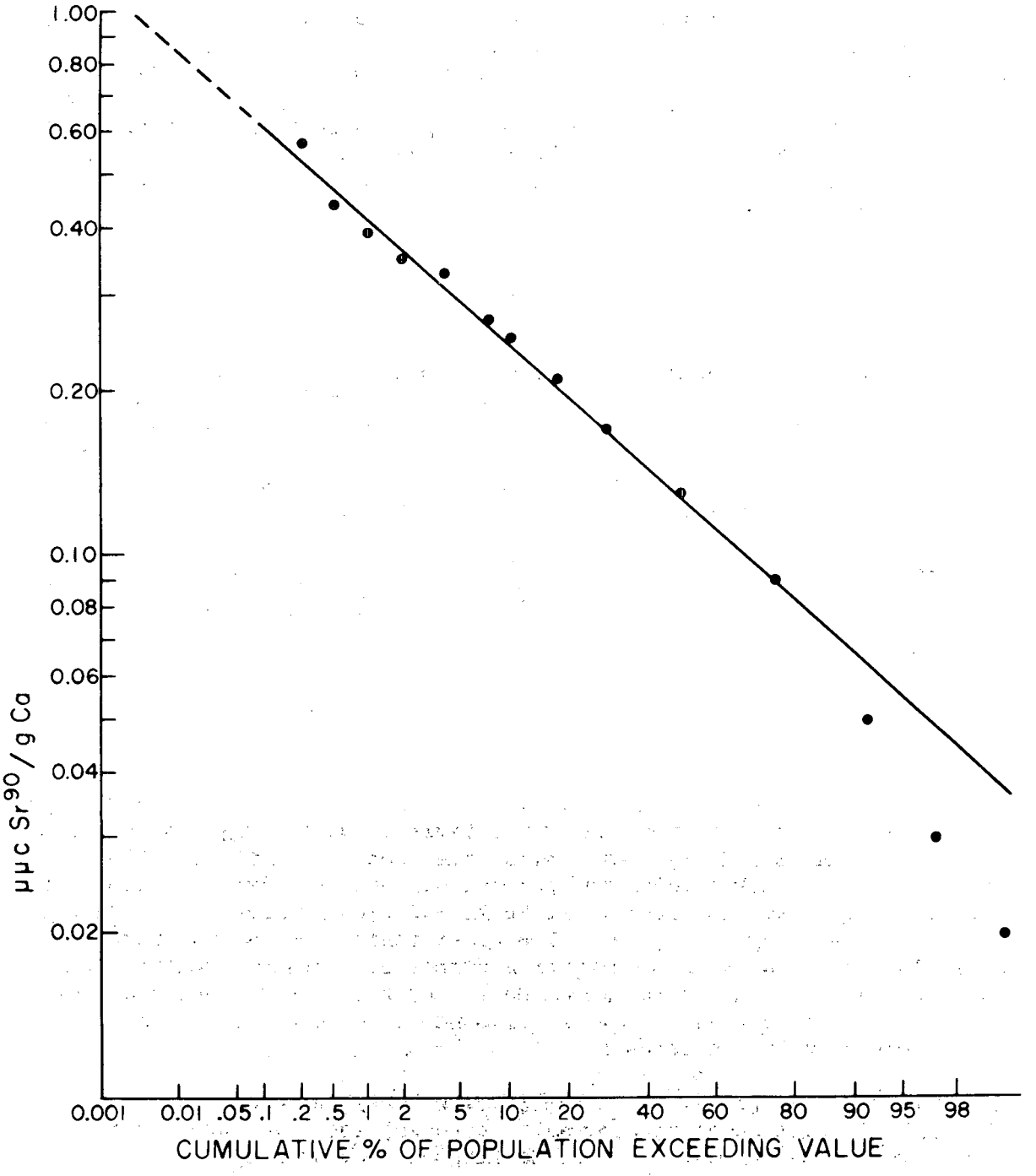

Figure 2. Distribution curve of N. Y. whole-skeleton samples plotted on probability-log normal scales. Average value of this population is 0.15 μμc Sr^{90}/g Ca.

populations that derive their food fr n local sources, there may be a wider dispersion of the Sr^{90} concentration. On the other hand, attempts to find greater dispersion have been unsuccessful (12). Isolated instances of high concentration have been found in the diets of the isolated Indian tribes of the upper Amazon and of inland Eskimos who live largely on caribou (12,4). There are undoubtedly other groups with a high Sr^{90} concentration in their food - groups such as vegetarians with a high whole-wheat component in their diet and inland rice farmers in the 30° to 60°N latitude zone - but these have not as yet been identified experimentally.

Conclusion

With the existing information on the mechanisms by which Sr^{90} passes from the site of injection into the stratosphere to the human population it is possible to estimate future bone concentrations for any specified injections. The major uncertainties are the fraction of debris that falls out locally in a surface burst and the variation in deposition with latitude. The uncertainties are least in the areas of maximum fallout (Temperate Zone latitudes and high rainfall) which also coincides with the maximum in the distribution of world population. It appears that the Sr^{90} concentration of the average adult in New York or London can be predicted using these data to better than a factor of two for most hypothetical conditions.

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CESIUM^{137} LEVELS IN UNITED STATES POWDERED MILK AND IN THE POPULATION*

E.C. Anderson, G.M. Ward,** J.Z. Holland,*** and W.H. Langham
Los Alamos Scientific Laboratory
University of California
Los Alamos, New Mexico

Introduction

Cs^{137} is a long-lived radionuclide formed in about 6% abudance in fast neutron fission of Pu^{239}, U^{235}, and U^{238} (1). It probably is secondary only to Sr^{90} as a potentially hazardous material in world-wide fallout from nuclear weapon tests. In event of nuclear war, local and tropospheric fallout of relatively short-and intermediate-lived radionuclides (e.g., I^{131}) may be limiting primarily over either Sr^{90} or Cs^{137}. The long radiological half-life of Cs^{137} affords a long integration time in the soil, and its chemical similarity to K (a required constituent of plants and animals) results in rapid entry into the biosphere. Since it emits both beta and gamma rays, it is potentially both an internal and an external radiation source. Its concentration in muscle results in radiation to the gonads, and its cumulation in the soil increases the general radiation background level. The accumulation of Cs^{137} on the earth's surface and in man, therefore, results in both a potential somatic and a genetic hazard to the general population. Since 1956, the Los Alamos Scientific Laboratory (LASL) has made periodic measurements of the Cs^{137} levels in the United States population and in area sam-

*Work performed under the auspices of the U.S. Atomic Energy Commission.
**On leave from the Animal Science Department, Colorado State University, Fort Collins.
*** Permanent address, Division of Biology and Medicine U.S. Atomic Energy Commission, Washington, D.C.

plings of the powdered milk supply. Reports of the primary data (2-7) and occasional preliminary interpretive articles (8-20) have been published from time to time. The present report is a final summary of the project. The milk measurements have been discontinued as of the time of resumption of nuclear weapon tests by the U.S.S.R. on September 1, 1961, because re-injection of fresh Cs^{137} will obscure the long-term trends of Cs^{137} in the cycle from production to foods and to man, which were of principal interest. A few measurements of people from the New Mexico area will continue to be made to determine rate and extent of increase of Cs^{137} body levels as a result of recent U.S.S.R. tests.

Methods

Sampling of United States Powdered Milk. Beginning in 1956, 50- to 100-lb. samples of powdered milk from major production areas in the U.S. were collected at weekly intervals and assayed for Cs^{137} and naturally-occurring K^{40}. During 1958-1959, the number of weekly sampling sites totaled 55, covering all major powdered milk producing areas of the U.S. and Canada. Since samples were obtained through normal channels from commercial milk-drying plants (which utilize only surplus milk), it was not possible to obtain samples from some States in which milk production does not exceed local requirements. Thus, the mountain and high plains areas (Nevada, Montana, Wyoming, Colorado, Nebraska, and Kansas) were not represented, and sampling points were few and intermittent in the Southeast. On July 1, 1960, the sampling network was curtailed by dropping the Canadian stations and cutting back the U.S. sampling to the 16 most widely distributed and dependable stations. In the fall and winter of 1960-1961, each of the 16 plants retained in the sampling network was visited to obtain detailed information on factors that may have bearing on Cs^{137} levels of milk produced in these specific areas. Dairying practices were evaluated with particular attention to number of cows, size of production area, production level, length of pasture season, type of pasture, type of hay, and amount of silage fed. Soil types and soil acidity were also investigated as well as rainfall patterns and milkshed uniformity. Information was gathered from discussions with plant managers and fieldmen, county agriculture agents, State university staff, and from some farm visits. In addition to detailed information for the 16 selected locations, general information was gathered for all areas that had supplied milk to the network. Some locations were visited and, for others, information was gathered by correspondence or discussion with knowledgeable people.

Sampling of the United States Population. Samplings of the general U.S. population were drawn from visitors to the Los Alamos Scientific

Laboratory who volunteered to be counted. The more intensive sampling of the New Mexico area was carried out by soliciting volunteers from the population of Los Alamos and surrounding areas. The yearly samplings averaged approximately 300 people and during each year represented in excess of 30 of the States of the Union. Cs^{137} and natural K^{40} were determined on all subjects.

Methods of Measuring and Reporting Cesium^{137} Levels in Milk and in People. Measurements of the gamma radioactivity of people and milk were made with the Los Alamos 4π liquid scintillation counter (21-24), which provided a geometrical efficiency of nearly 100%. The energy resolution of the detector was adequate to provide accurate simultaneous measurements of K^{40} (gamma energy, 1.46 Mev) and Cs^{137} (gamma energy, 0.662 Mev). Collateral measurements of the gamma spectra of people and milk with a 9 in. NaI crystal spectrometer (23,25,26) showed that, except for milk samples collected immediately following weapon tests in Nevada in 1957 and 1958 (Operations PLUMBBOB and HARDTACK II), Cs^{137} and K^{40} were the only gamma radioactivities present. Standard counting procedure involved a counting time of 200 secs. per sample or subject. The precision and stability of the detector were such that the K^{40} and Cs^{137} in a 50-lb. milk sample could be determined with an average error (1σ) of about 2 and 5% respectively, and the K^{40} and Cs^{137} levels in an average human subject could be determined with an average error (1σ) of about 3% and 6 pc/g K, respectively. These error estimates are based on the experimentally observed precision of 197 measurements on 2 control milk samples covering a 4-yr. period and on 398 measurements of 4 control subjects (3). The absolute error for K has been shown to be the same as the precision on the basis of comparison of the counting results with measurements on the same subjects by K^{42} isotope dilution and by comparison of averages for given age groups with those obtained by NaI crystal spectrometry. The absolute error of the Cs^{137} may be considerably larger (perhaps as much as 20 to 30% as judged by interlaboratory comparisons (18). This uncertainty is a systematic one affecting all results equally, and the relative values are not affected.

Measurements of Cs^{137} levels in milk are reported in pc Cs^{137}/g K, which may be converted to average pc Cs^{137}/l of liquid milk by multiplying by 1.4, the average g of K in 1 liter of milk. Cs^{137} levels in people are reported in pc Cs^{137}/g body potassium. These values may be converted to average adult body burden of Cs^{137} (in pc) by multiplying by 140 g, the average body potassium content of the standard 70-kg man.

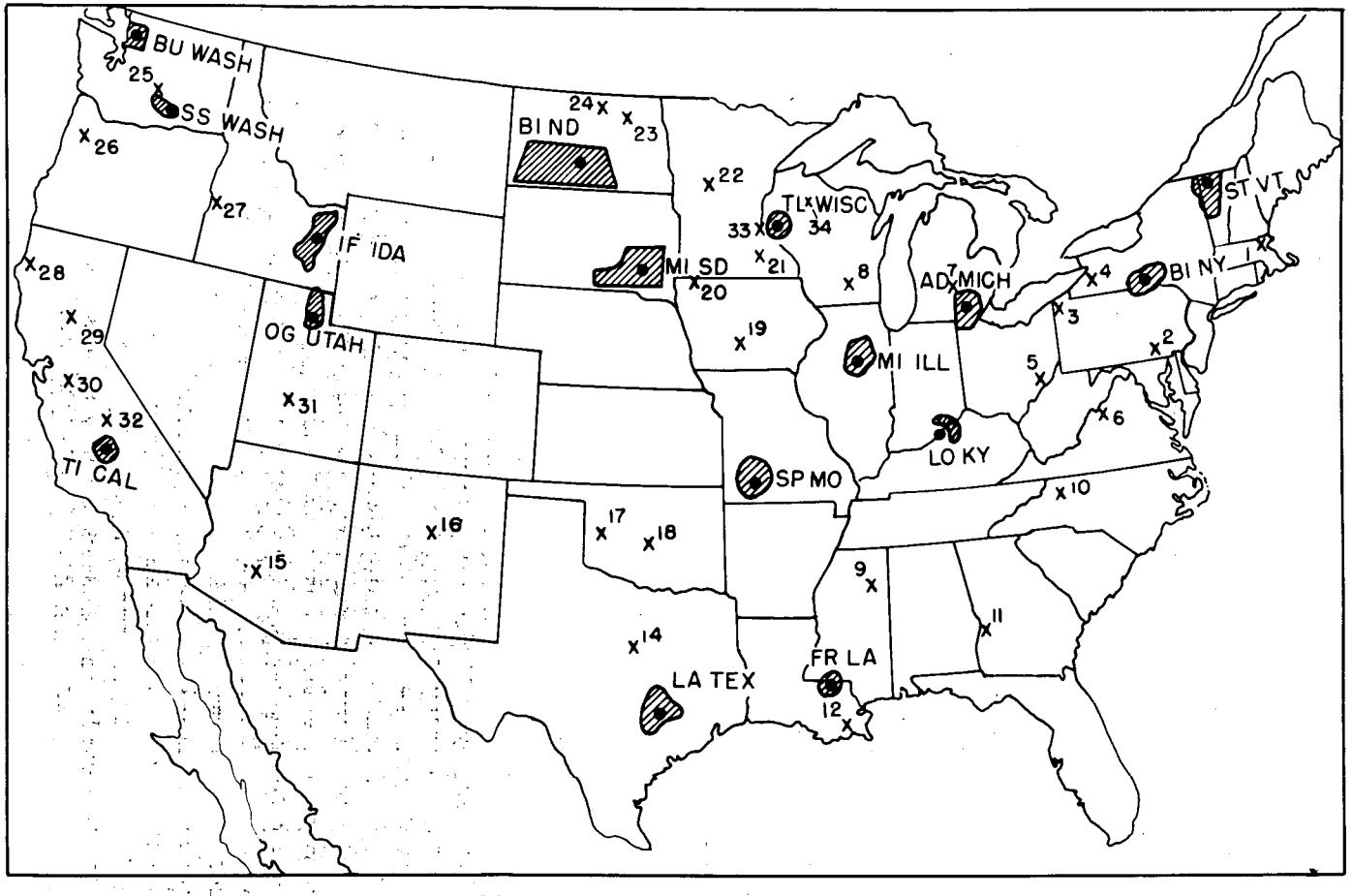
Results and Discussion

Cs^{137} Levels in United States Powdered Milk

Survey of Milk Stations. --Fig. 1 shows the U.S. sampling network. The cross hatched areas represent the 16 stations that were retained after July 1, 1960, and the approximate milkshed area from which milk is supplied to the processing plants. The numbered crosses show locations of stations sampled prior to July 1960. Table 1 gives the designation and specific location of the supply point for each of the stations shown in Fig. 1.

In Table 2 is tabulated some of the information obtained from surveying the locations which were retained in the LASL sampling network. The 16 locations retained after July 1, 1960, are listed as ranked according to internal homogeneity of dairy production practices of farms supplying the plant. Considerations involved in this ranking were seasonal variations in milk supply, soil types, and rainfall patterns within the milkshed, which in turn produced variations in feeding practices. Most of the field survey data are of necessity highly subjective, but they give some feeling for the wide variability in agricultural conditions, milk production levels, and dairying practices among the milk stations and throughout the U.S.

Time-Trend in U.S. Powdered Milk. -- Smoothed time graphs of Cs^{137} levels (expressed as pc/g K) in U.S. powdered milk covering the period from the first of 1957 through the third quarter of 1961 are shown in Fig. 2. These curves were derived from analyses of the weekly samplings from the various stations. For comparison, the stations have been grouped arbitrarily into regions on the basis of apparent similarity in meteorological conditions, dairy feeding practices, and pasturing season. Data from some of the stations shown in Fig. 1 and listed in Table 1 were omitted. In these cases, the stations were, for various reasons, too unreliable to give sufficiently consistent data to make presentation worthwhile. The time-trends presented show a regular seasonal fluctuation at all stations, with maxima generally occurring in the spring and minima occurring in the late summer. This pattern is reminiscent of seasonal variation of fission product concentration in surface air and of fallout rate, with perhap some indication of damping of the oscillations and shifts in maxima and minima, possibly as a result of seasonal alterations in the times animals were on fresh pasture in the spring and on stored feed during the non-pasture season. Table 3 shows quarterly averages of Cs^{137} levels for these same stations over the same time period. These data show that the time of quarterly maxima was shifted toward the third and fourth quarters



LASL MILK SAMPLING POINTS

CROSSHATCHED AREAS INDICATE THE LOCATION AND EXTENT OF MILK SHED FOR SAMPLING POINTS RETAINED AFTER JULY 1, 1960

x INDICATES POINTS WHICH WERE FORMERLY PART OF SAMPLING NETWORK

Figure 1. Locations of supply points in the powdered milk sampling network.

TABLE 1. STATION DESIGNATION AND SPECIFIC LOCATION OF SUPPLY POINT FOR ALL POWDERED MILK STATIONS

Station No. or Designation	Station Supply Point	Station No. or Designation	Station Supply Point
Prior to July 1, 1960		After July 1, 1960	
1	Andover, Massachusetts	TI CAL	Tipton, California
2	York, Pennsylvania	IF IDA	Idaho Falls, Idaho
3	Springboro, Pennsylvania	MI ILL	Minonk, Illinois
4	Little Valley, New York	LO KY	Louisville, Kentucky
5	Marietta, Ohio	FR LA	Franklinton, Louisiana
6	Harrisonburg, Virginia	AD MICH	Adrian, Michigan
7	Lansing, Michigan	SP MO	Springfield, Missouri
8	Deerfield, Wisconsin	BI NY	Binghamton, New York
9	Aberdeen, Mississippi	BI ND	Bismarck, North Dakota
10	Wilkesboro, North Carolina	MI SD	Mitchell, South Dakota
11	Columbus, Georgia	LA TEX	La Grange, Texas
12	New Orleans, Louisiana	OG UTAH	Ogden, Utah
14	Fort Worth, Texas	ST VT	St. Albans, Vermont
15	Glendale, Arizona	SS WASH	Sunnyside, Washington
16	Albuquerque, New Mexico	BU WASH	Burlington, Washington
17	Elk City, Oklahoma	TL WISC	Turtle Lake, Wisconsin
18	Norman, Oklahoma		
19	Des Moines, Iowa		
20	Sibley, Iowa		
21	Claremont, Minnesota		
22	Bertha, Minnesota		
23	Cando, North Dakota		
24	Bottineau, North Dakota		
25	Ellensburg, Washington		
26	McMinnville, Oregon		
27	Payette, Idaho		
28	Fernbridge, California		
29	Willows, California		
30	Newman, California		
31	Monroe, Utah		
32	Fresno, California		

TABLE 2. SUMMARY OF FIELD SURVEY OF AGRICULTURAL CONDITIONS AND DAIRYING PRACTICES AT POWDERED MILK SAMPLING STATIONS

Location (Ranked According to Homogeneity)	Powder from Grade A Milk (per cent)	No. Herds	No. Cows	Acreage Required per Cow	Milk per Acre per Day (lbs)	Start of Pasture	Old Sod (per cent)	Soil pH	Rainfall 4-yr Ave. (in.)	Irrigation
Tipton, California	95	115	13,500	1	35	April 1	10	>7.0	8	++
Louisville, Kentucky	100	175	4,000	3	8	May 1	30	6.5	41	--
Minonk, Illinois	5	800	5,600	3	5	April 15	50	6.0	33	--
Franklinton, Louisiana	100	1700	42,000	3	5	All year	30	5.7	64	--
La Grange, Texas	100	300	12,000	4	4	All year	30	7.0	46	--
Lansing, Michigan	100	225	6,800	3	10	May 1	20	6.5	30	--
Adrian, Michigan	85	1300	16,000	3	10	May 1				
Sunnyside, Washington	20	280	3,000	2	13	April 1	50	-	8	++
Burlington, Washington	90	800	36,000	3	12	April 1	30	6.0	31	--
Idaho Falls, Idaho	50	1500	12,000	4	6	May 1	20	>7.0	7	++
Mitchell, South Dakota	5	440	5,000	6	3	May 10	70	-	22	--
Bismarck, North Dakota	0*	4000	20,000	12	1	May 1	90	6.8	13	--
Binghamton, New York	100	1100	28,000	4	6	May 1	40	5.7	41	--
St. Albans, Vermont	100	950	28,000	6	6	May 15	50	5.5	32	--
Stillwater, Minnesota	70	1600	30,000	5	5	May 15	50	5.7	24	--
Turtle Lake, Wisconsin	60	670	14,000			May 15			24	
Ogden, Utah	90	500	14,000	3	12	May 1	10	>7.0	13	++
Springfield, Missouri	20	3200	45,000	1	5	April 1	80	5.3	43	--

*Dried buttermilk only.

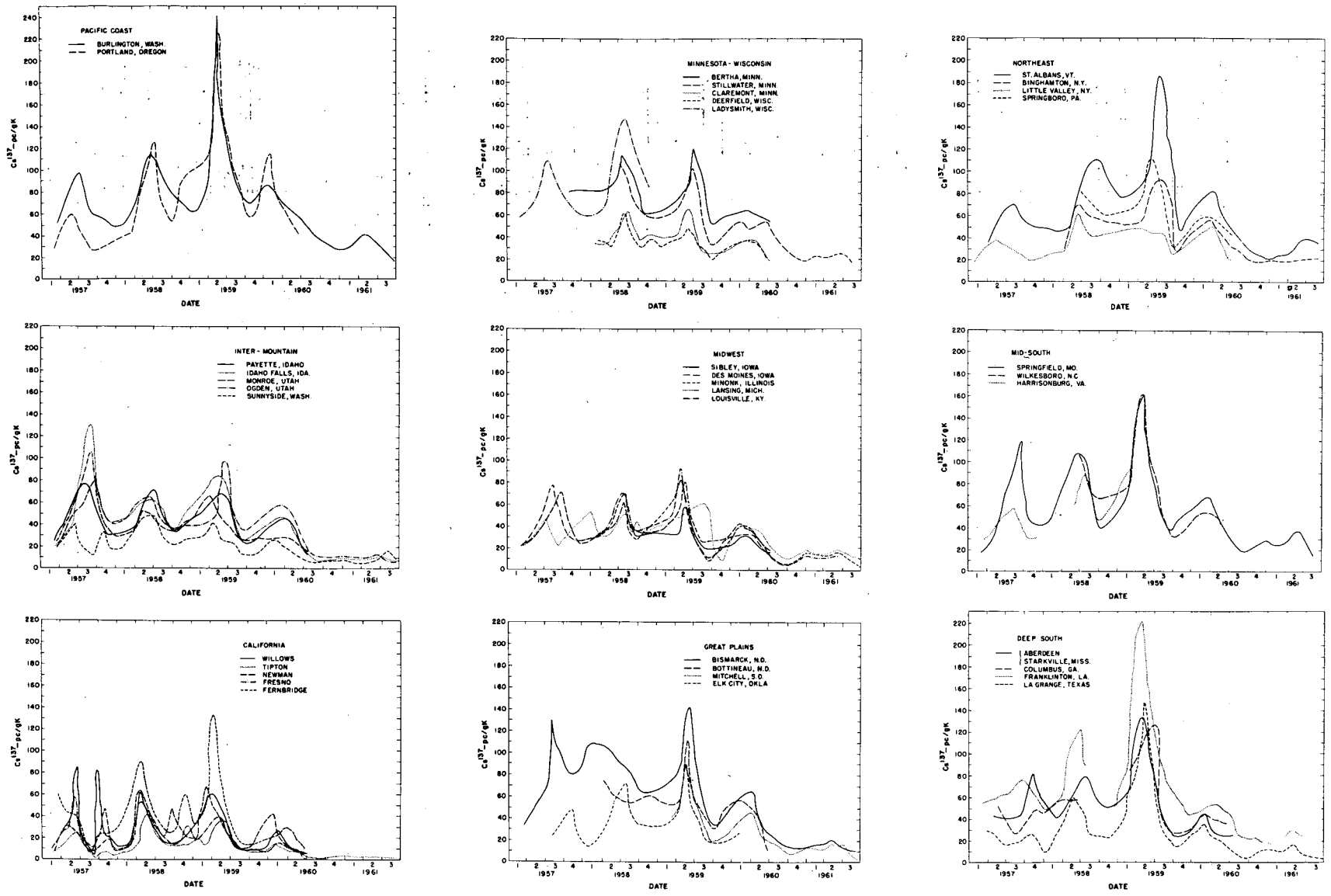


Figure 2. Time-trends in the Cs^{137} levels or U. S. powdered milk.

of the years 1957-1958 at those stations in a general northerly and easterly direction from the Nevada Test Site. This shift can be explained on the basis of tropospheric fallout from Nevada test operations. After cessation of tests in November 1958, peak quarterly average milk levels at all stations were confined almost entirely to the first and second quarters of the year. Except for one station, the maximum quarterly average occurred in the first quarter of 1960 and shifted again to the second quarter in 1961. This shift may have been related to the relative importance of current grazing and winter feeding of highly contaminated forage grown during the spring and summer of 1959, when the contamination level was relatively high.

Quarterly averages during 1961 are rather interesting in that they show practically no change from quarter to quarter. This observation suggests that both spring fallout rate and contamination level of winter feed had dropped to a point where other factors were beginning to contribute significantly to milk levels, possibly Cs^{137} uptake from the soil or from the root mat in those areas where sodded pastures may have provided a high percentage of spring and summer grazing. Undoubtedly, the time variation in Cs^{137} milk levels is influenced by a multiplicity of factors involving, among other things, time and place of weapon tests, rate of fallout and its seasonal fluctuations, alternation of lot and pasture feeding between winter and spring months, and such agronomic factors as soil and pasture type, with the latter factors being significant only when the fallout rate is relatively low. The extremely rapid response of Cs^{137} levels in milk to change in fallout rate is highly suggestive that the observed Cs^{137} levels in the biosphere during the period of observation were predominantly dependent on fallout rate and its direct contamination of vegetation and not on integral surface deposition level operating through true ecological uptake via the roots of plants. Empirical correlation of milk levels with available measurements of fallout and with dairy feeding practices would strengthen this argument and provide a basis for empirical determination of effective time lags and quantitative coefficients for purposes of predicting contamination levels in milk as a result of more weapon tests.

Correlation of Milk Levels with Fallout Rate and with Dairy Feeding Practices. --The best available estimates of short-term average fallout rates were obtained from the measurements of Sr^{90} deposition in pot and funnel collectors. Because of mismatching of fallout and milk sampling stations, different sample intervals, different area-averaging characteristics, and other factors, it was necessary to deal with averages of several samples of each type over a long period of time to bring out systematic relationships. The only area of the U.S. for which adequate data of both types were

TABLE 3. QUARTERLY AVERAGES OF CESIUM^{137} LEVELS (pc/g K) IN POWDERED MILK FROM VARIOUS LOCATIONS IN THE UNITED STATES AS A FUNCTION OF YEARS

Region and Location	1957 Quarter				1958 Quarter				1959 Quarter				1960 Quarter				1961 Quarter			Rainfall 4-yr Ave. (in.)
	1	2	3	4	1	2	3	4	1	2	3	4	1	2	3	4	1	2	3	
California																				
Tipton, California	12	16	12	6	9	33	17	14	18	33	15	9	13	8	3	3	2	4	2	8
Fresno, California	18	29	15	13	16	44	22	22	24	32	14	9	11	7	-	-	-	-	-	11
Newman, California	11	30	16	25	17	49	30	26	39	31	13	18	20	7	-	-	-	-	-	12
Willows, California	22	36	28	21	14	39	20	20	41	41	9	10	20	8	-	-	-	-	-	19
Fernbridge, California	60	44	17	26	43	64	29	41	-	80	19	24	28	20	-	-	-	-	-	45
Pacific Coast																				
Portland, Oregon	29	55	38	31	38	91	78	78	98	152	81	66	82	57	-	-	-	-	-	38
Burlington, Washington	60	80	71	54	50	85	77	80	72	162	94	78	78	64	43	33	6	11	7	31
Intermountain																				
Ogden, Utah	29	47	72	49	46	58	48	40	39	60	53	43	54	44	14	10	11	12	9	13
Monroe, Utah	23	33	51	35	31	42	37	43	51	50	32	24	26	26	-	-	-	-	-	8
Payette, Idaho	32	39	47	43	34	55	48	38	51	70	36	27	43	34	-	-	-	-	-	12
Idaho Falls, Idaho	25	46	78	55	47	57	53	48	64	73	39	34	43	35	12	11	9	13	9	7
Sunnyside, Washington	20	39	17	26	19	41	31	23	28	32	17	16	22	14	8	7	6	11	7	8
Great Plains																				
Bismarck, North Dakota	36	55	84	84	87	87	85	65	70	97	66	43	55	43	21	16	16	19	14	13
Bottineau, North Dakota	-	-	-	-	-	62	57	53	51	70	48	44	54	36	-	-	-	-	-	14
Mitchell, South Dakota	-	-	-	-	-	-	-	-	-	64	37	26	36	30	17	11	12	14	10	22
Elk City, Oklahoma	-	-	26	30	27	39	47	36	35	65	27	21	25	-	-	-	-	-	-	31

TABLE 3 (continued)

Region and Location	1957 Quarter				1958 Quarter				1959 Quarter				1960 Quarter				1961 Quarter			Rainfall 4-yr Ave.
	1	2	3	4	1	2	3	4	1	2	3	4	1	2	3	4	1	2	3	(in.)
Minnesota-Wisconsin																				
Bertha, Minnesota	-	-	47	83	-	82	100	64	68	79	90	56	-	-	-	-	-	-	-	24
Stillwater, Minnesota	-	-	-	-	-	-	86	60	60	79	60	40	51	50	37	26	-	-	-	24
Ladysmith, Wisconsin	59	71	90	78	59	76	124	-	-	-	-	77	-	-	-	-	-	-	-	31
Turtle Lake, Wisconsin (Stillwater, Minnesota)	-	-	-	-	-	-	86	50	60	79	60	40	51	50	37	26	23	24	25	24
Claremont, Minnesota	-	-	-	-	33	33	50	37	40	51	34	28	35	33	-	-	-	-	-	27
Deerfield, Wisconsin	-	-	-	-	35	34	46	32	36	40	32	27	33	34	-	-	-	-	-	33
Midwest																				
Sibley, Iowa	-	-	-	-	-	-	34	29	31	41	27	18	26	23	-	-	-	-	-	27
Des Moines, Iowa	19	28	51	33	27	34	44	32	40	62	27	25	38	29	-	-	-	-	-	32
Minonk, Illinois	-	-	-	-	-	36	47	35	55	65	20	21	35	28	10	8	14	14	8	33
Lansing, Michigan (Adrian, Michigan)	25	32	34	37	46	35	43	36	41	48	55	20	37	33	16	14	15	15	11	30
Louisville, Kentucky	22	26	55	31	27	44	43	36	41	59	21	18	30	22	10	8	12	12	-	41
Midsouth and South																				
Springfield, Missouri	20	34	66	54	42	77	80	42	59	125	57	50	56	47	24	24	22	32	22	43
Wilkesboro, North Carolina	-	-	-	-	-	-	78	64	76	130	67	38	50	-	-	-	-	-	-	48
Aberdeen, Mississippi	-	43	40	66	46	63	62	52	71	104	44	27	40	26	-	-	-	-	-	54
Franklinton, Louisiana	56	59	73	59	53	90	108	59	86	168	86	51	47	47	28	21	21	27	30	64
La Grange, Texas	25	24	18	25	23	40	27	23	57	89	25	20	30	19	9	8	10	12	5	46
Northeast																				
Springboro, Pennsylvania	-	-	-	-	-	-	71	62	68	89	62	42	56	51	-	-	-	-	-	45
Binghamton, New York	-	-	-	-	-	45	57	54	53	64	73	40	50	43	31	35	19	21	20	41
St. Albans, Vermont	-	44	62	55	46	53	85	96	80	101	128	63	79	62	39	25	25	29	31	32

available was the Midwest for the period 1957-1961.

The milk stations chosen were Claremont, Minn., Sibley and Des Moines, Ia., Deerfield, Wis., Minonk, Ill., Adrian, Mich., and Louisville, Ky. Fallout pot sampling stations were operated during all or part of the period at Vermillion, S. Dak., Columbia, Mo., Lemont, Ill., Green Bay, Wis., and Louisville, Ky. The actual fallout at the milk stations was assumed to be given by the fallout at the pot stations multiplied by the ratio of the average precipitation the milkshed (based on data furnished by the U.S. Weather Bureau) to the average precipitation at the pot stations.

During some of 1957 and 1958, the monthly milk averages were invalidated by the presence of short-lived radioactivities (principally I^{131} and Ba^{140} - La^{140}). These samples were not included in the subsequent correlation analysis.

Retention of radioactivity at various stages in the ecological cycle would be expected to introduce delays into the response of milk Cs^{137} to changes in fallout rate. The available information on transfer coefficients, exchange rates, and half-times was inadequate to permit construction of a theoretical model a priori. Therefore, various empirical equations were tested in which milk radioactivity in a given month was assumed to depend on fallout rate for several months preceding the month of production, progressively decreasing weights being given to the earlier months. The best correlation was obtained with the equation,

$$\bar{F}_m = (4\,F_m + 3\,F_{m-1} + 2\,F_{m-2} + F_{m-3})/10$$

where F_m is the observed fallout rate for month m, and $\bar{F}_m$ is the weighted time-average fallout rate used for correlation with the milk level.

A graph of the resulting correlation is shown on the left in Fig. 3 in which the monthly average Cs^{137}/K ratio of milk is plotted against fallout adjusted according to the above equation using only those months during which the cows were on pasture. The aberrant points are for the months of May 1958, 1959, and 1960, and suggest that "memory" of winter feed level may be a significant factor. The fallout equation was, therefore, modified by substituting,

$$F_m' = (F_{May} + 2\,F_{June} + F_{July})/4$$

for any F_m where m is an off-pasture month, since during such months the cows are presumed to be eating forage grown in the preceding May-July. Data for all months of the year can now be included with

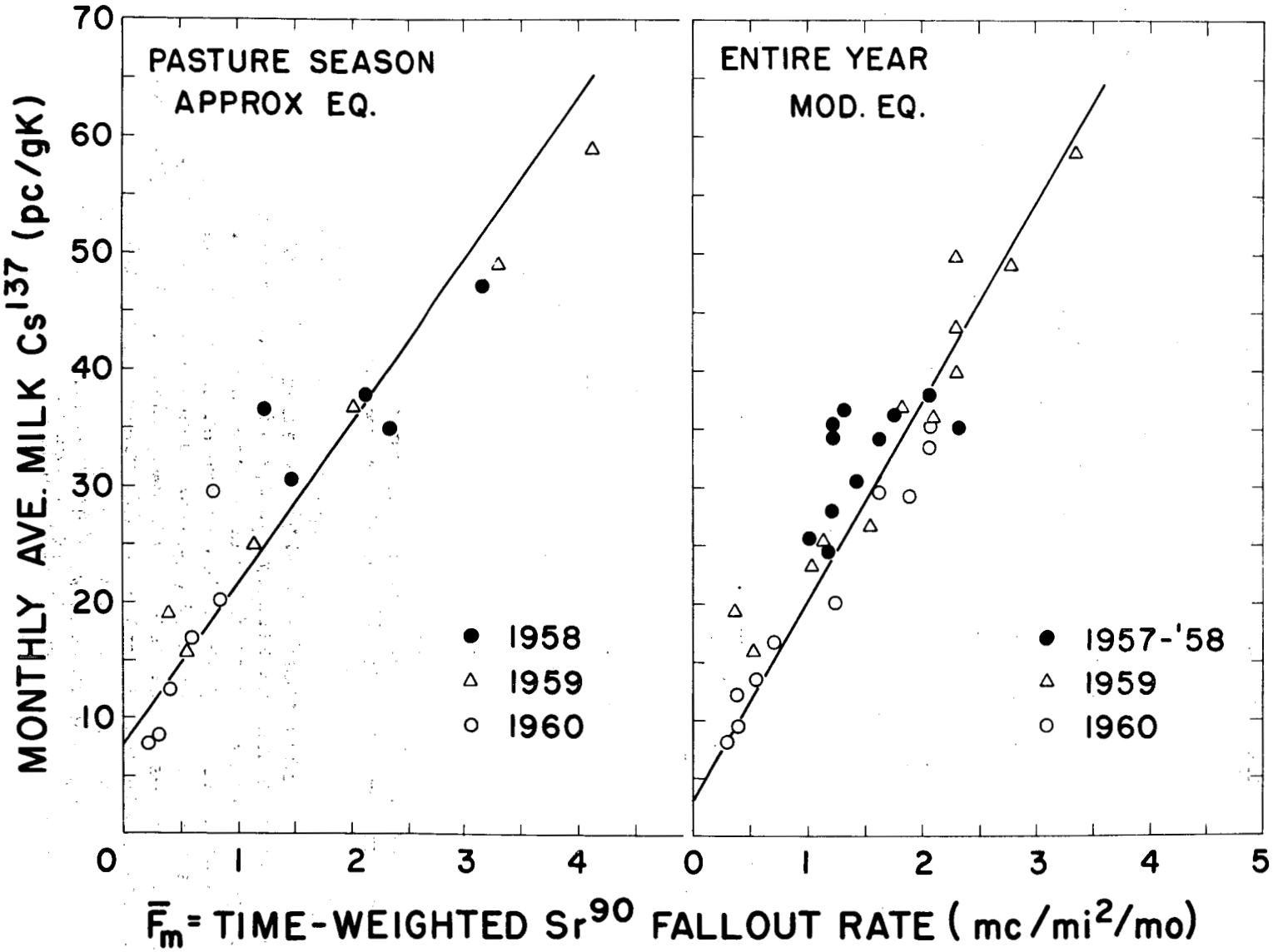


Figure 3. Correlation between monthly average Cs^{137} milk level and the time-weighted fallout rate.

the result shown on the right in Fig. 3. The estimated regression formula is:

$$M_m = 3 + 17\,\bar{F}_m$$

for M_m in pc Cs^{137}/g K, and $\bar{F}_m$ in mc Sr^{90}/mi.^{2}/mo.

A rather important observation that may be made from these results is that there is little or no evidence of uptake of Cs^{137} from the soil. Within the precision of the data, the line extrapolates essentially to zero Cs^{137} at zero average monthly fallout rate.

Figure 4 shows a comparison of the observed milk values (solid line) with the values calculated from the above regression equation (open triangles) and with the fallout rate (dashed line). The fit of the calculated line to the observed data is surprisingly good, considering the complexity of the situation. Attempts to apply the same empirical equation to other regions were not highly successful, perhaps because of insufficient milk data and mismatching of milk and fallout sampling stations.

Geographic Distribution of Cs^{137} Milk Levels. --The raw average of Cs^{137}/K measurements for all milk samples collected from January 1957 through December 1959 is plotted at each station location in Fig. 5. There is no obvious direct relation between this pattern and the integral Sr^{90} surface deposition pattern derived from 1959 soil data reported by Alexander et al. (27). Evidently average Cs^{137} levels in milk for the period 1957-1959 were primarily influenced by factors other than integral surface deposition level. Cs^{137} milk levels were relatively low in California and the lower Southwest, intermediate in the Corn Belt and upper Southwest, and high across the northern border, the Pacific Northwest, and the East Coast.

Since fallout rate and surface deposition level are assumed to be more or less proportional to rainfall, it was of interest to consider the geographic distribution of average Cs^{137} levels in milk with the average annual rainfall in the various parts of the country. Fig. 6 shows a map of the ratio of average Cs^{137} /K levels for 1957-1959 (the period prior to the test moratorium) to the average annual rainfall at each station for the same period. Milk data from too few sampling stations were available to permit a similar correlation for the period during which the test moratorium was in effect. Broad similarities to the pattern of fallout per inch of precipitation emerge, however, from the earlier data (27). The highest ratios were in the Great Basin area with relatively low and uniform ratios along the Pacific Coast, Gulf of Mexico, and south Atlantic Coast. The over-all range is about a factor of 10, which is in general

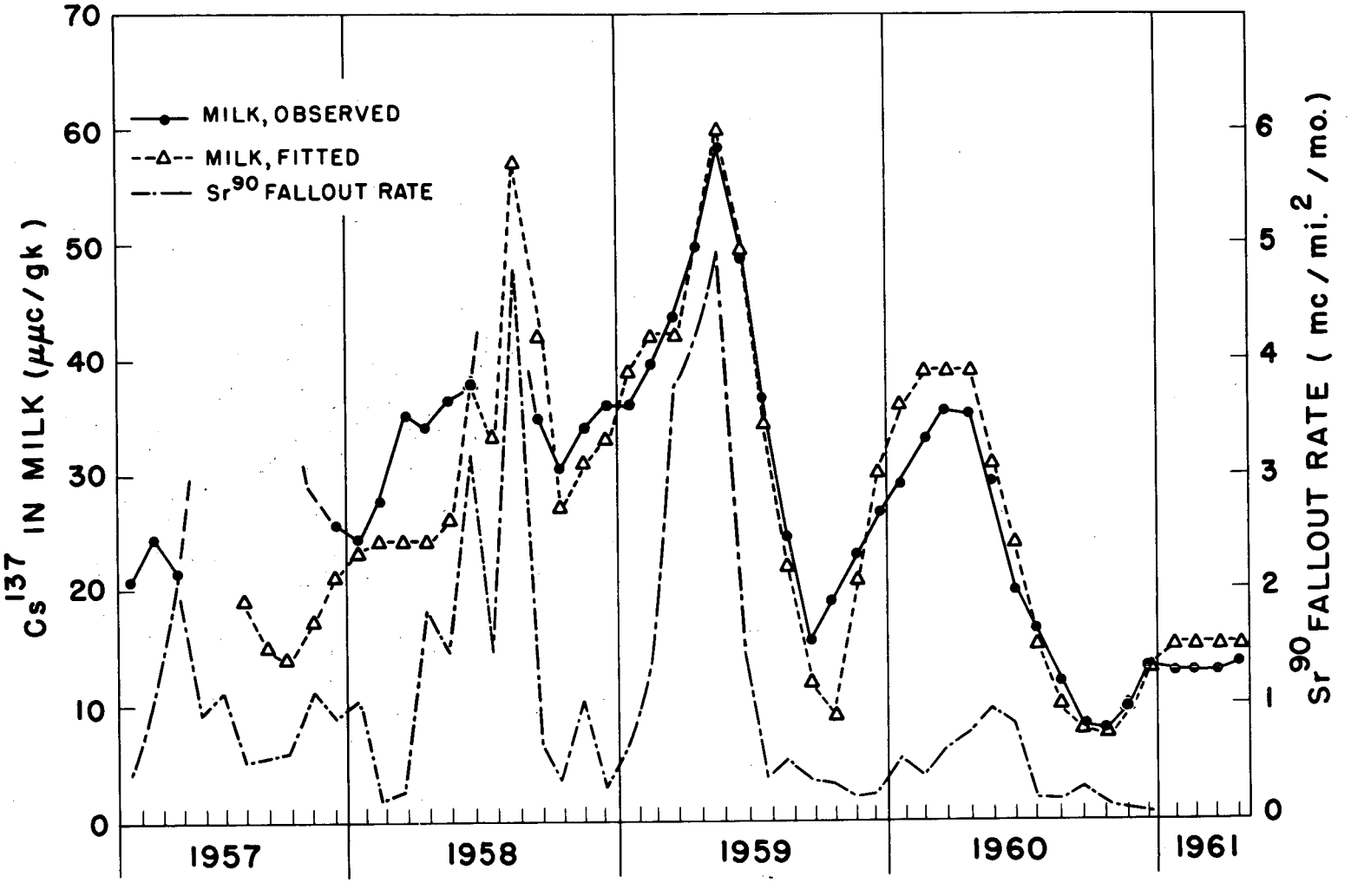


Figure 4. Comparison of observed Cs^{137} monthly milk levels with levels calculated from time-weighted fallout rate.

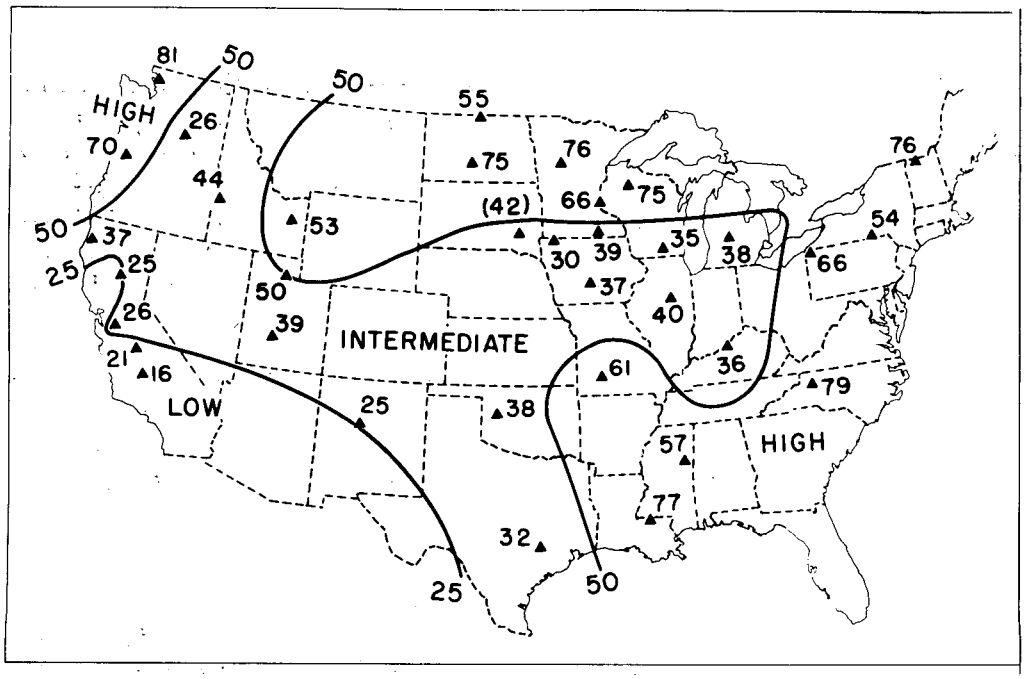
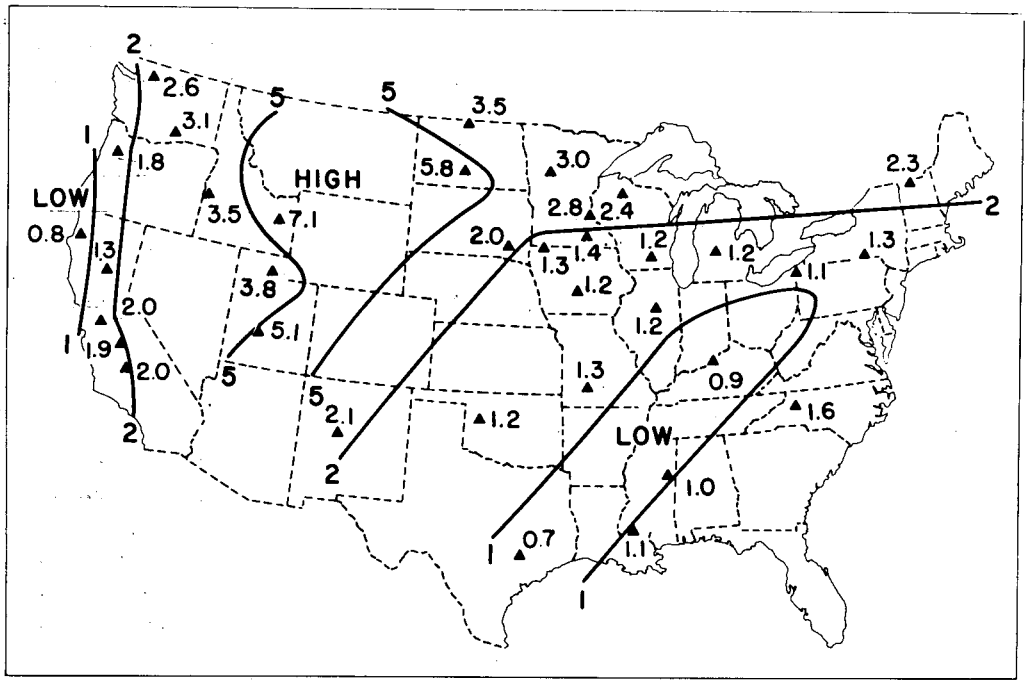


Figure 5. Geographic distribution of 1957-1959 average Cs^{137} milk levels.



Figures 6. Geographic variation of the ratio of average Cs^{137} milk level to average annual rainfall.

agreement with the range for fallout concentration (a factor of ~8) derived from Sr^{90} data. Despite a poor geographic coverage of sampling stations, it appears that the highest milk ratios tended to occur farther north than the highest fallout ratios and that the milk/precipitation ratio showed a minimum in the Mississippi-Ohio Valley. The pattern of areas of high and low milk activity per inch of precipitation has a striking resemblance to the pattern of frequency of continental and maritime air masses (28).

Attempts to deduce quantitative relationships between annual rainfall and yearly average Cs^{137} levels in milk have not been particularly successful. Crude correlations, however, did permit groupings of certain stations into regions possessing some degree of geographical unity from year to year (29).

Cs^{137} Levels in the U.S. Population

Time-Trend in New Mexico Residents. -- Because of proximity to the human counter, residents of northern New Mexico provided a relatively representative sampling of a reasonable homogeneous population group. The large number of quarterly samplings of this area provided an excellent opportunity to study the time-trend of Cs^{137} levels in a population relatively unperturbed by sampling and other problems. Fig. 7 shows a plot of the quarterly average Cs^{137} levels (log scale) in New Mexico residents as a function of date from the first quarter of 1956 through the third quarter of 1961. These data provide a basis for a number of interesting observations.

The Cs^{137} levels in the population group show seasonal fluctuations as do fallout rate and Cs^{137} levels in milk. Prior to 1959, yearly maxima and minima in Cs^{137} levels in people, however, were out of phase with those of the fallout rate by about 6 to 8 months. Maxima in the population group occurred in the fall and minima in the spring, which were essentially the opposite of the fallout rate. During the period January 1956 through the first quarter of 1959, the fall maxima in the population levels, therefore, appeared to be a manifestation of the high contamination levels occurring in the previous spring and perhaps to some degree (as a result of winter feeding of beef and dairy stock) the spring prior to that. This phenomenon may be explained on the basis of average agronomic, metabolic, and food-distribution and marketing time lags. Furthermore, changes in the population level were not as abrupt as changes in fallout rate, which may be explained also by damping effects of the above factors.

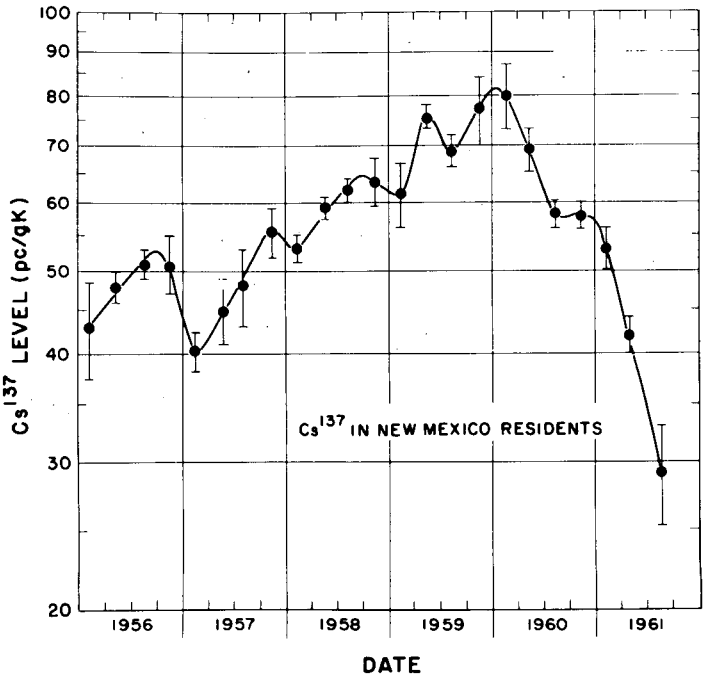


Figure 7. Quarterly average Cs^{137} levels in New Mexico residents for the period 1956-1961.

Beginning in 1959, perturbations in the population Cs^{137} level occurred which may have been due, in part, to the unusually precipitatous decline of fallout rates following the spring maximum of that year, resulting from the large U.S.S.R. test series at immediately preceding the test moratorium as northerly latitudes in the fall of 1958. The gross result was an early maximum in about May, which dropped to a minimum in about August. In 1960, the maximum occurred in the first quarter of the year, which was the time of previous minima. This may be a result of the drop in food contamination levels from those of the stored winter foods reflecting the heavy fallout of early 1959 to the lower levels of the fresh spring milk and produce of 1960.

The lack of a well defined peak during the last quarter of the year is consistent with this picture since the contribution of spring 1960 stratospheric fallout was too small to overcome the longer-term downward trend resulting from gradual exhaustion of stored foods grown during previous seasons of high fallout rate. The data indicate that the relation between the fallout rate in the previous growing season, reflected in winter food contamination levels, and the fallout rate in the new growing season, as well as the changes

in fallout rate during the growing season itself, can have a determining effect on the phase of the seasonal variation of Cs^{137} in people.

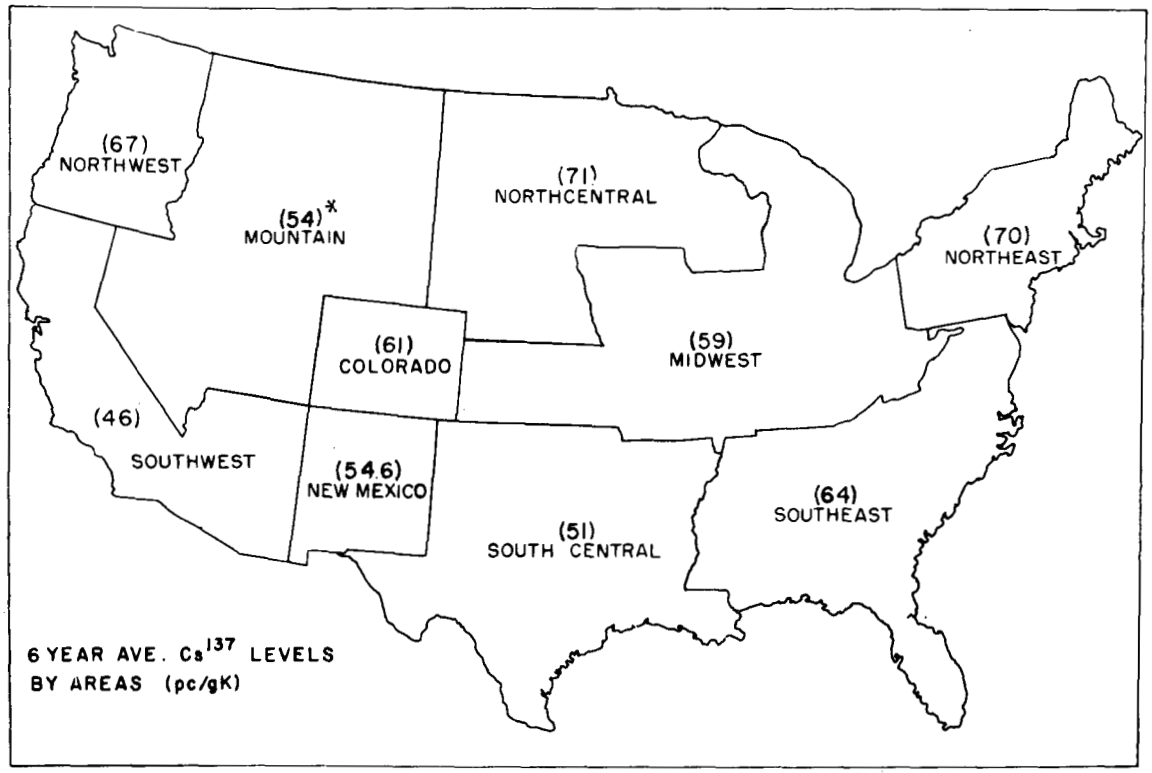
From the peak value in 1956, Cs^{137} levels in the New Mexico population group dropped with an apparent effective half-time of about 9 months, from the 1960 maximum with an apparent half-time of about 12 months, and during 1961 with a half-time of about 8 months (average 10 months). Effective half-times of Cs^{137} in military personnel residing in Japan-Korea and Washington, D.C., during 1958-1959 were estimated to be about 7 and 14 months, respectively (30), and a value of about 15 months was obtained for a group of German subjects (31) during 1960. The average half-time in New Mexico residents is considerably longer than the biological half-time of Cs^{137} in man (4 to 5 months) but agrees with the fallout half-time (10 months) assumed by Stebbins (32) for fission products injected into the equatorial stratosphere below 70,000 ft. This again suggests that the population Cs^{137} levels follow the longer-term (non-seasonal) trends of fallout rate.

It will not be possible to determine whether the 1961 Cs^{137} levels in New Mexico residents would have continued to drop with a 10-month average half-time had weapons testing not been resumed. One might conjecture, however, that a very slight indication of leveling off might have occurred during the last quarter, followed by a continuing downward trend, eventually assuming a flatter slope approaching that of stratospheric fallout of fission debris injected between 70,000 and 100,000 ft. /about 20 months (32)/, and finally assuming a still flatter slope indicating the point at which true ecological uptake via root absorption from the soil begins to be the dominant factor in the food contamination level.

Although the data in Fig. 7 were collected for New Mexico residents only, they are in general qualitative agreement with less extensive data from the Argonne National Laboratory in Chicago (33-35), the United Kingdom Atomic Energy Research Establishment in Harwell (36), the Walter Reed Army Research Institute in Washington, D.C. (30), and the U.S. Army Medical Research Unit in Landstuhl, Germany (31). In general, these data suggest peak levels in the fall prior to 1959, in some cases a double peak after the October 1958 test operations, and generally rapidly dropping levels during 1960-1961. Quantitatively, the relative height of peaks and their exact times of occurrence vary. This may be expected in view of the great differences in climatic conditions, fallout rate, dairy practices, food distribution and marketing systems, and other factors at such widely separated areas as New Mexico, Washington, D.C., Chicago, England, and Germany.

Geographic Distribution of Average Cs^{137} Levels in the United States Population. -- Sufficient data are available to estimate 1956-1961 average concentrations of Cs^{137} in people from various parts of the U.S. For purposes of this comparison, the U.S. was divided into 10 regions (as shown in Fig. 8). The method of division was determined by climatic and geographic uniformity, modified by the number of subjects available. Certain states, such as New Mexico and Colorado, were sufficiently represented that they were treated as statistically significant units. The "Mountain" area was so poorly represented that an average value could not be derived except by assuming the average was that of the surrounding areas of Colorado, New Mexico, and the general Southwest. The "Northwest" was of questionable homogeneity because of the extreme differences in rainfall on opposite sides of the Cascade Range. Similar objections apply to other areas, in some cases moderate by selectivity in the actual sources of subjects (e.g., the Californians were largely from the southern, dry part of the State). Furthermore, since samples were drawn from visitors to Los Alamos and hence represented unusually mobile subgroups, the relative averages may not correspond to the true population-weighted conditions for the various regions. It is possible also that variations in source and composition of diet occur with variations in family income. In this event, the extent to which local fallout conditions are reflected in man may vary with general economic level of the various areas of the country. The data are presented with these reservations. The 6-year (1956-1961) average Cs^{137} levels (in pc/g K) for the various regions (not population-weighted) are shown in Fig. 8. The most outstanding feature of the data is the extremely small variation over the entire U.S. The averages varied only from 48 to 71 pc Cs^{137}/g K.

Similar data on geographic distribution of Cs^{137} levels in the U.S. population were reported by the Walter Reed Army Institute of Research (30). The data were averaged over different periods (July 1958-June 1959; July 1959-June 1960) and over arbitrarily chosen geographic regions, which do not correspond exactly with the areas shown in Fig. 8. The regions were sufficiently similar, however, to provide some general comparison of their regional averages for July 1958-June 1960 with regional averages of the present data (Table 4) over approximately the same period (January 1958-December 1960). The two sets of data are in relatively good agreement both qualitatively and quantitatively, considering the differences in regions and times over which the averages were drawn. The regional average levels from the Los Alamos results are 10 to 15% higher than the Walter Reed averages for the same general areas. The latter data also show the surprisingly small variation throughout the entire country. The observed narrow spread throughout the U.S. may be partly due to sampling bias (mobile population,



SUBDIVISION OF THE U.S.
POPULATION SAMPLE BY AREAS

Figure 8. Regional distribution of 6-year average Cs^{137} levels in the United States population.

above average income), but undoubtedly the country's rather wide food distribution system would have a significant equalizing effect on the various regional levels when compared with each other and with a national average.

Table 4 summarizes the yearly relative average concentration of Cs^{137} in the population for the past 6 years. To eliminate effects of a secular change, the New Mexico average (the best defined statistically) was taken as unity for each year and the averages for other regions referred to it. For reference, the actual New Mexico averages (in pc/g K) are given in the last line. The number of subjects is given in parentheses below each average.

The consistency of the ratios for a given area over the 6-year period adds confidence that the differences are real. In addition, the general agreement of the yearly pattern of the ratios with that of milk levels, rainfall, and primary fallout is as expected. New Mexico was one of the lowest areas and California-Arizona the lowest. The highest areas were the Northeast, Northcentral, Northwest, and other areas intermediate -- in general qualitative agreement with milk results. Combining the factors in Table 4 weighted by population

TABLE 4. COMPARISON OF AVERAGE CESIUM^{137} LEVELS IN SUBJECTS FROM VARIOUS PARTS OF THE UNITED STATES (1956-1961)

Area	Units Relative to New Mexico Averages[a]						
	1956	1957	1958	1959	1960	1961[b]	Average
Southeast	1.10[c] (23)	1.05 (21)	1.10 (21)	1.18 (16)	1.46 (13)	1.18 (13)	1.18 (107)
Northeast	1.33 (19)	1.27 (31)	1.22 (40)	1.22 (21)	1.52 (13)	1.15 (26)	1.29 (150)
Southcentral	0.81 (14)	1.00 (11)	1.05 (35)	0.93 (33)	0.93 (13)	0.90 (24)	0.94 (130)
Midwest	1.01 (45)	1.16 (45)	1.17 (47)	1.10 (32)	1.14 (47)	0.92 (39)	1.08 (255)
Northcentral	1.49 (3)	1.13 (12)	1.37 (16)	1.19 (6)	1.19 (15)	1.40 (34)	1.30 (86)
Southwest	0.74 (17)	0.89 (20)	0.85 (39)	0.87 (20)	0.99 (16)	0.67 (19)	0.84 (131)
Colorado	0.81 (6)	1.30 (28)	1.00 (14)	1.07 (47)	1.05 (6)	1.07 (9)	1.12 (110)
Northwest	1.10 (5)	1.38 (15)	1.23 (12)	1.14 (9)	1.30 (9)	1.18 (10)	1.22 (60)
Mountain	Too few subjects					0.86 (10)	
New Mexico[a] (in pc/g K)	49.6 (143)	44.8 (103)	58.1 (223)	72.3 (81)	61.6 (143)	41.2 (81)	54.6 (774)

[a]New Mexico average = 1.00 for each year.

[b]Through September

[c]Number of subjects given in parentheses below ratio.

gives a 6-year (1956-1961) average factor of 1.16 for the ratio of United States average to New Mexico average.

Frequency Distribution of Cesium^{137} Levels in the United States Population. --Table 5 summarizes the frequency distribution of Cs^{137} levels in New Mexico subjects for the years 1956 through the third quarter of 1961. "Adult" subjects only (arbitrarily defined as all subjects weighing more than 40 kg) were included to eliminate possibility of any error because of a systematic variation of counter efficiency in a weight range where *in vivo* calibration was not feasible. (Actually, no difference was observed between the yearly averages for the two weight groups greater and less than 40 kg.) Reported in the table are the cumulative number of subjects having less than a given Cs^{137} level and the corresponding cumulative fraction of the population sample. The number of subjects varied from 215 (1958) to 81 (1959 and 1961). These data are plotted in Fig. 9 on linear probability paper. This method of presentation expresses normal frequency distribution as a straight line with slope proportional to the standard deviation; the steeper the slope, the larger the standard deviation. The curves for 1956 and 1959 appeared normal, and 1957 and 1961 were not significantly abnormal. Data for both 1958 and 1960 however, appear to show a significant non-normal trend at high Cs^{137} levels. The number of subjects whose Cs^{137} levels exceeded 100 pc/g K were 5 and 4, respectively, compared with expectations of 0.4 and 0.25 from normal curves.

Table 6 and Fig. 10 give the same data for all U.S. adults measured during this period. In this case, a deviation from normal distribution was detectable in all years except 1956 and 1961 and was especially pronounced in 1959. The more pronounced abnormality in 1959 may have resulted from inclusion of subjects from areas of higher fallout related to the unusual conditions posed by the 1958 U.S.S.R. test series at far northerly latitudes. The latter data are replotted in Fig. 11 on log probability paper; here a log-normal distribution would give a straight line. The fit of the data to straight lines is somewhat better. The year 1959 still shows an upward deviation at high Cs^{137} levels, and all curves now show a significant drop at low Cs^{137} values (i.e., too many subjects show very low Cs^{137} values). Neither the normal nor the log-normal curve is, therefore, completely successful in describing the data.

For purposes of predicting maximum probable radiation doses to individuals, the region of the frequency curve beyond 99.8% may be of importance. However, since these persons are encountered at the rate of only 2/1000, a large body of data is required to validate predictions in this range. Pooling of all data from Los Alamos, the Army Medical Research Unit, Europe (31), and the Walter Reed Army Institute of Research (30) would give a total approaching 10,000

TABLE 5. DISTRIBUTION OF CESIUM^{137} LEVELS IN NEW MEXICO ADULTS (1956-1961[a])

Cs^{137} (pc/g K)	1956		1957		1958		1959		1960		1961	
	Cumulative	Fraction	Cumulative	Fraction	Cumulative	Fraction	Cumulative	Fraction	Cumulative	Fraction	Cumulative	Fraction
0	0	0	1	0.0099	0	0	0	0	0	0	2	0.026
10	1	0.0070	5	0.0495	1	0.0046	0	0	1	0.0079	4	0.052
20	15	0.106	19	0.188	4	0.0186	0	0	3	0.0238	21	0.259
30	49	0.345	41	0.406	14	0.0652	0	0	8	0.0634	40	0.494
40	79	0.548	70	0.693	70	0.325	4	0.0494	33	0.262	64	0.790
50	107	0.754	84	0.832	129	0.600	19	0.234	61	0.484	72	0.889
60	125	0.880	92	0.911	180	0.836	37	0.456	96	0.761	76	0.936
70	133	0.936	98	0.970	202	0.940	59	0.729	115	0.912	78	0.962
80	140	0.985	99	0.980	207	0.964	68	0.839	121	0.960	80	0.986
90	141	0.992	100	0.990	211	0.981	77	0.951	122	0.967	81	1.000
100	142	1.000	101	1.000	213	0.990	80	0.989	123	0.975		
110					214	0.995	81	1.000	125	0.991		
120					215	1.000			125	0.991		
130									125	0.991		
140									126	1.000		
150												
160												

[a]Through September 1961.

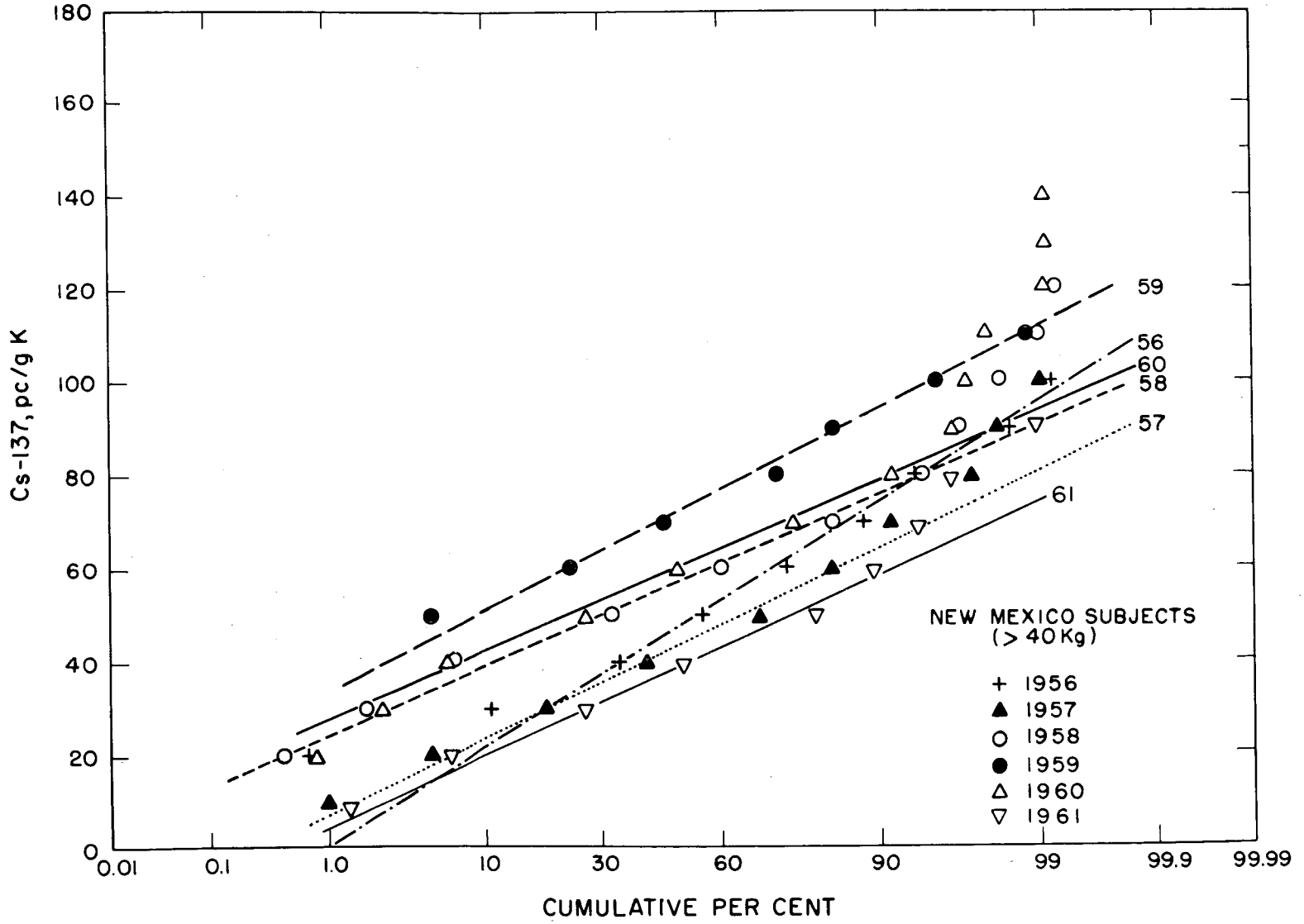


Figure 9. Normal frequency distribution plot of Cs^{137} levels in New Mexico residents.

TABLE 6. DISTRIBUTION OF CESIUM^{137} LEVELS IN ALL UNITED STATES ADULTS (1956-1961[a])

Cs^{137}	1956		1957		1958		1959		1960		1961	
(pc/g K)	Cumulative	Fraction	Cumulative	Fraction	Cumulative	Fraction	Cumulative	Fraction	Cumulative	Fraction	Cumulative	Fraction
0	1	0.0038	1	0.003	0	0	0	0	0	0	4	0.015
10	6	0.0229	8	0.027	1	0.0024	0	0	0	0	15	0.056
20	33	0.126	28	0.094	4	0.0092	1	0.0033	3	0.019	67	0.250
30	85	0.325	81	0.272	24	0.0551	6	0.0199	9	0.036	134	0.500
40	147	0.561	160	0.536	116	0.266	21	0.069	41	0.156	192	0.716
50	197	0.753	223	0.748	222	0.511	68	0.225	92	0.363	227	0.848
60	225	0.860	253	0.849	318	0.731	126	0.417	154	0.586	246	0.918
70	248	0.947	276	0.926	381	0.876	201	0.666	200	0.761	260	0.971
80	259	0.990	286	0.959	407	0.936	240	0.795	225	0.849	264	0.986
90	261	0.996	294	0.986	424	0.975	274	0.907	240	0.913	268	1.000
100	262	1.000	298	1.000	428	0.985	289	0.956	249	0.948		
110					432	0.994	294	0.974	257	0.978		
120					435	1.000	297	0.983	261	0.992		
130							298	0.986	262	0.996		
140							300	0.994	263	1.000		
150							300	0.994				
160							301	0.997				
170							302	1.000				
180												

[a] Through September 1961.

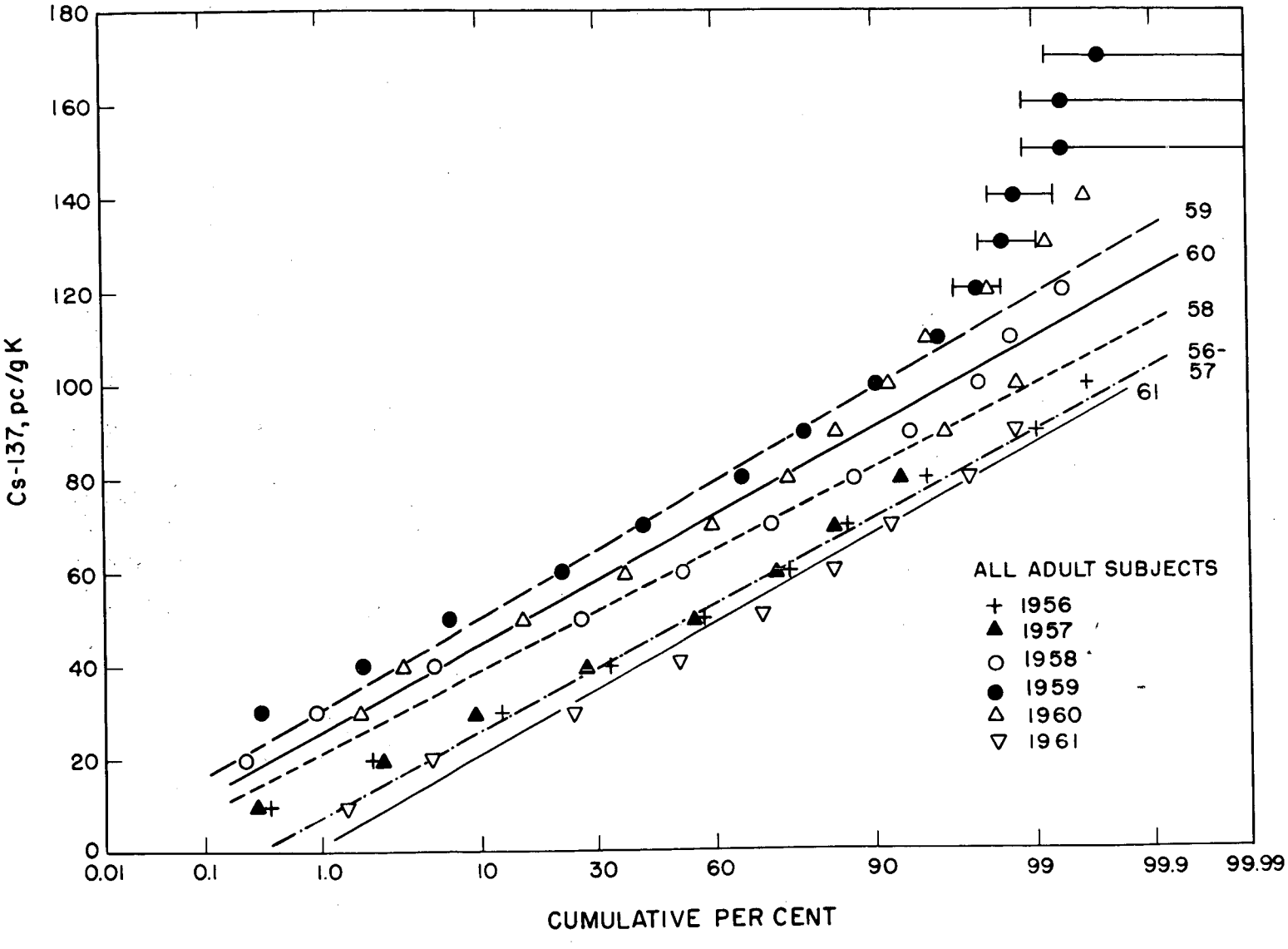


Figure 10. Normal frequency distribution plot of Cs^{137} levels in adult United States residents.

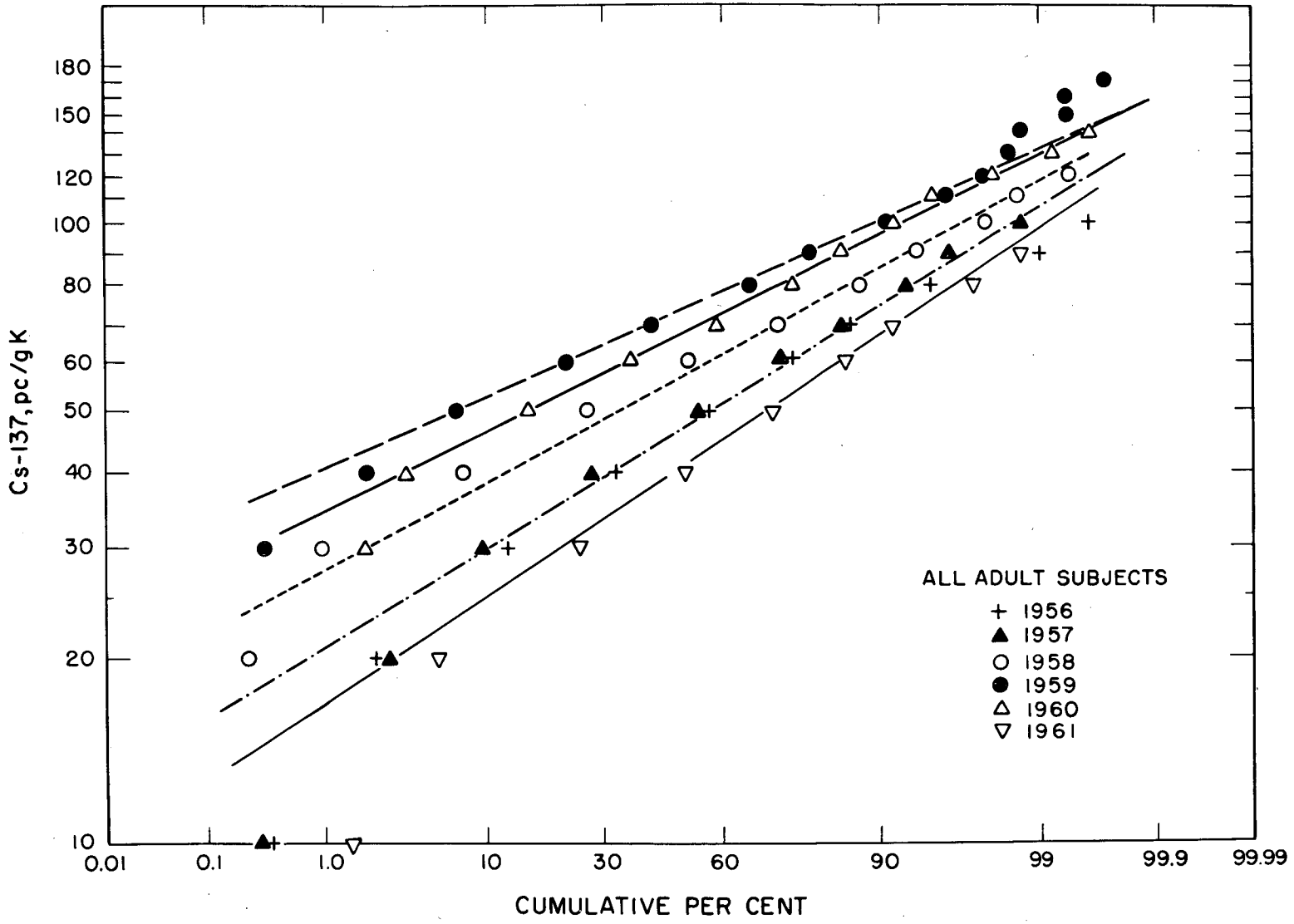


Figure 11. Log-normal frequency distribution plot of Cs^{137} levels in United States residents.

subjects in which one might expect to find 20 examples with Cs^{137} body burdens in the region of the 99.8 per cent probability level. It is possible at present only to say, on the basis of extrapolation, that the chances of a U.S. resident having a Cs^{137} body burden that is 3 times the mean may be only about 1 in 1000 to 1 in 10,000.

Interrelationships Among Cs^{137} Fallout Rate, Population Levels, Milk Levels, and Levels in the Balance of the Diet. -- From the pot collection data summarized by the AEC's Health and Safety Laboratory (37), it was possible to derive a U.S. quarterly average fallout rate for Sr^{90} for the period 1956 through the third quarter of 1961. Assuming no fractionation of Sr^{90} and Cs^{137}, multiplication of the Sr^{90} averages by 1.7 gives the U.S. average Cs^{137} fallout rate. These data are shown in the lower curve of Fig. 12 in which a smoothed curve is drawn through the points representing the quarterly average fallout rate expressed as mc $Cs^{137}/mi.^2$ Although some variation in time of yearly peak fallout rate occurred as a result of Nevada testing, the average time for the 6-year period was about mid-April. It was possible to compare Cs^{137} milk levels for the same period with fallout rate by population-weighting the yearly average milk levels when grouped according to the areas shown in Fig. 8. When population-weighted in this crude way, the weighted average was approximately 1.09 times the non-weighted average. A smoothed curve drawn through the quarterly average milk levels multiplied by 1.09 is shown by the upper curve in Fig. 12. A striking correlation between milk Cs^{137} levels and fallout rate is apparent. The average time of peak milk concentration is about mid-May, showing approximately a 1-month delay between peak milk level and peak fallout rate. Following the peaks in mid-May, the Cs^{137} concentration in milk dropped with an average half-time of about 9 months until the beginning of the next rise, which usually began around the first of each year.

The relationship between the population-weighted quarterly average Cs^{137} levels in the U.S. population and the population-weighted quarterly average milk levels is shown in Fig. 13. The points for the population curve were established by multiplying the quarterly average Cs^{137} levels for New Mexico residents by the population-weighting factor derived from the yearly average data given in Table 4. A smoothed curve through the points shows a strong correlation between Cs^{137} levels in the population and in milk and, consequently, a strong correlation between fallout rate and Cs^{137} levels in the population. Other than for an early peak occurring in 1959 (which may be related to early fallout from the U.S.S.R. 1958 test series), the average time of occurrence of the yearly peak concentration in people is about the first of November. There is then a delay of approximately 6 months between the peak level in people and the peak level in milk and about a 7-month delay between the peak in people and the peak in the fallout rate. The data show also that,

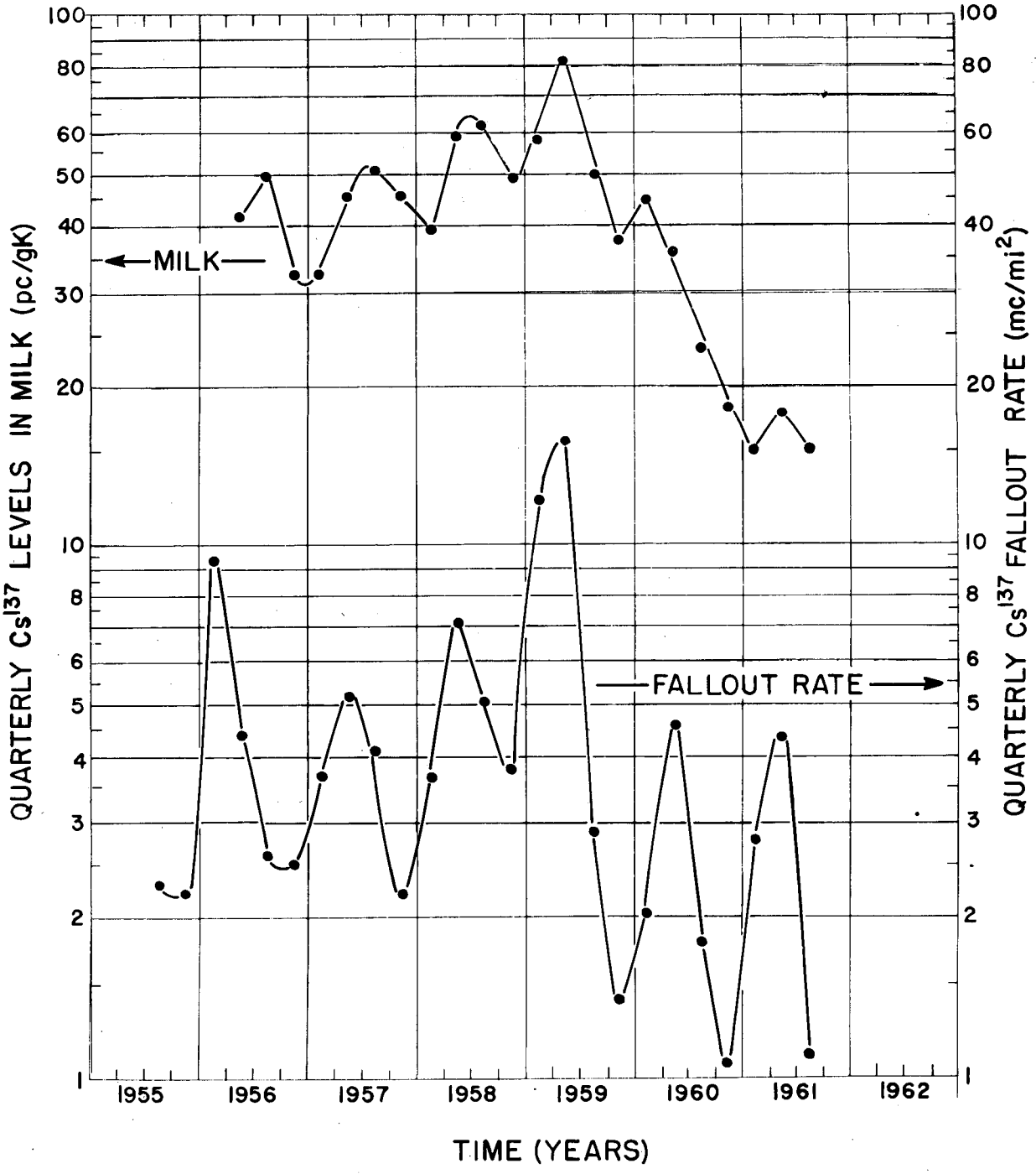


Figure 12. Relationship between quarterly average fallout rate and population-weighted quarterly average Cs^{137} levels in milk.

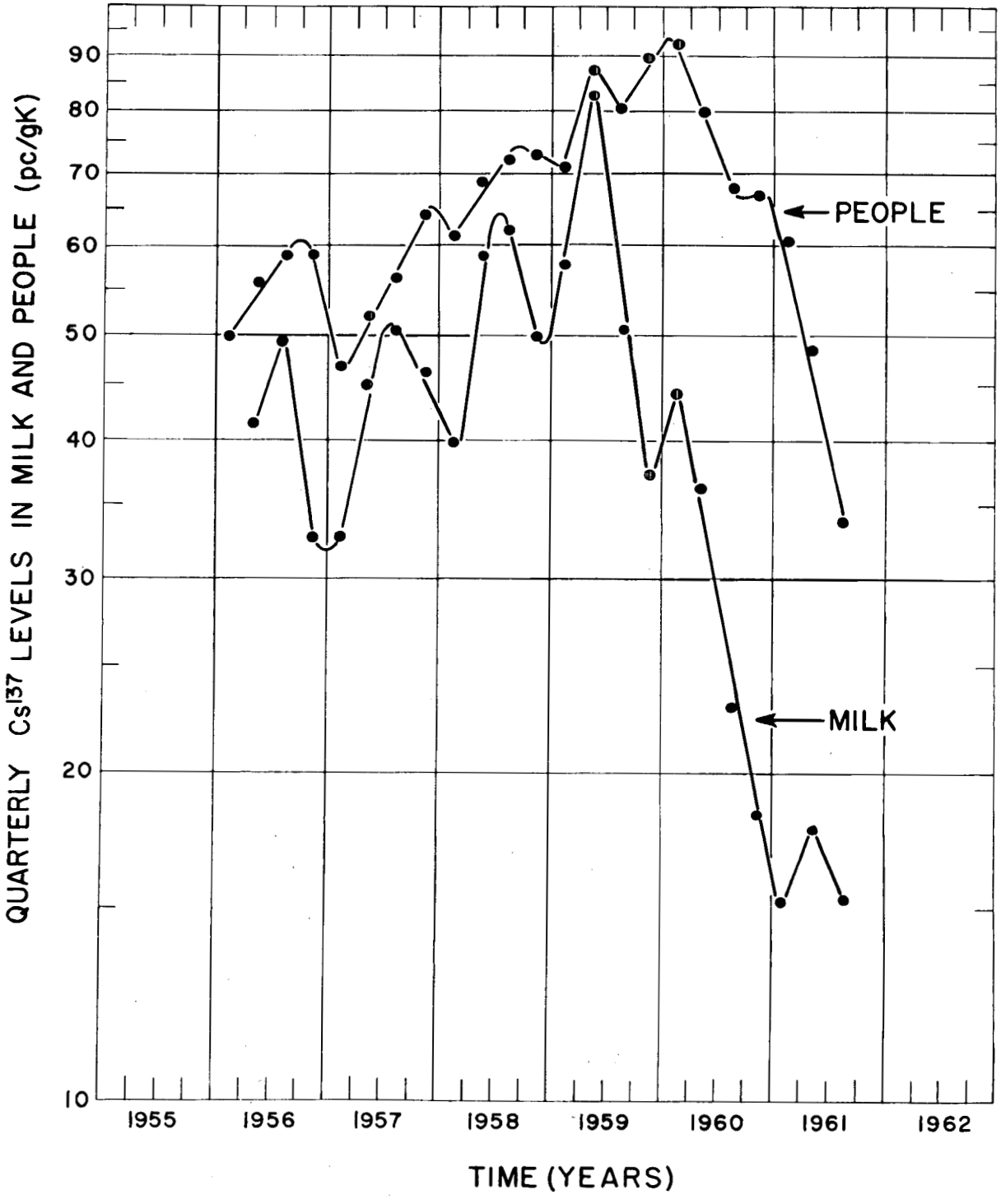


Figure 13. Relationship between population-weighted quarterly average Cs^{137} levels in the United States population and in milk.

with one exception (1958), the rate of fall of the Cs^{137} levels in people (average half-time 10 months) was approximately the same as that of milk. It is interesting to compare this value with the estimate of 10 months for the rate of fallout of fission products injected into the equatorial stratosphere below 70,000 ft. (32). Although this simple trend is not so evident in the fallout rate (Fig. 12) because of much more pronounced seasonal fluctuations, it appears that by 1961 the half-time for the remaining stratospheric fallout had become longer so that a slowing down of the decline of Cs^{137} in people might have been expected in the absence of resumed testing.

Undoubtedly, the delay time between peak concentrations in people and in milk is related to distribution and consumption time lags in foods other than milk. Essentially no data are available on Cs^{137} levels in foods other than milk. It is possible, however, to derive indirectly the average Cs^{137} levels in the balance of the diet from the levels in people and in milk if the discrimination factor for Cs^{137} over K in going from diet to man is known. A value for the discrimination factor may be derived from the data shown in Fig. 14, which shows a plot of Cs^{137}/g K in people against Cs^{137}/g K in milk for

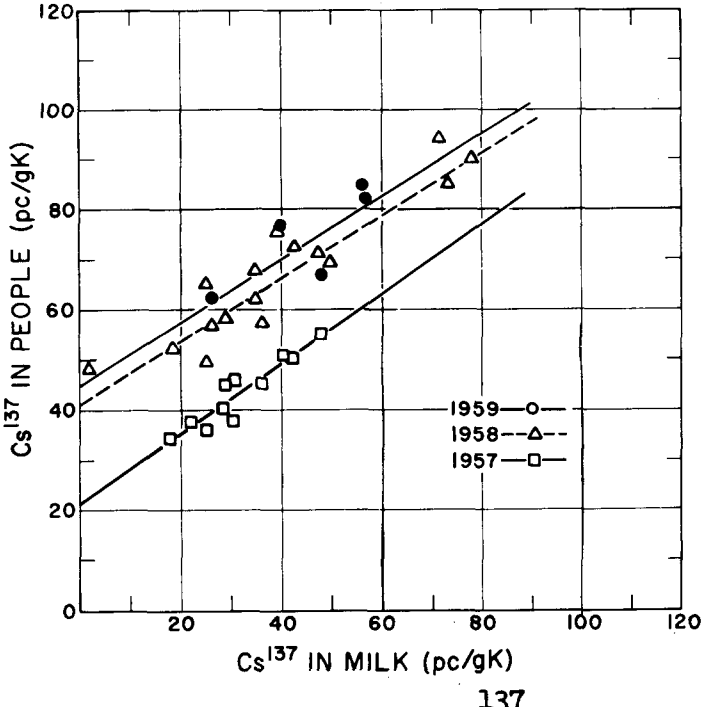


Figure 14. Correlation between average Cs^{137} levles in people and milk from the same area.

TABLE 7. RELATIVE CONTRIBUTION OF MILK TO YEARLY AVERAGE CESIUM^{137} LEVELS IN PEOPLE AND THE AVERAGE CONCENTRATION OF CESIUM^{137} IN THE BALANCE OF THE DIET

Year	Milk Concentration (pc/g K)	Concentration in People (pc/g K)	Calc. Conc. in Rest of Diet* (pc/g K)	Contribution of Cs^{137} Milk to Body Cs** (per cent)
1956	41.0	56.0	27.6	49.8
1957	43.7	54.6	24.4	54.4
1958	53.0	68.7	32.1	52.4
1959	57.1	82.2	42.5	47.2
1960	30.6	71.5	49.7	29.1
1961	16.0	47.9	36.3	22.8
Average	40.2	63.5	35.4	42.6

*Concentration in milk x 1.7 x 0.4 = milk contribution to people. Concentration in people minus milk contribution = contribution of balance of diet. Contribution by balance of diet ÷ 1.7 x 0.6 = concentration in balance of diet.

**Milk contribution x 100 ÷ concentration in people = per cent contribution by milk.

those areas and years (1957-1959) where adequate matching of population and milk measurements was possible. The average slope for the three lines is 0.65 which, when divided by that fraction of dietary K derived from milk (0.4), gives ~ 1.7 as the average discrimination factor. The intercept of each line should be the pc Cs^{137}/g K in people that was derived from the balance of the diet in each of the 3 years.

If it is assumed that the discrimination factor in going from the balance of the diet may be calculated from the yearly average levels in milk and in people and the fraction of dietary K (0.6) derived from the balance of the diet. Results of the calculation are shown in Table 7. These data show that the relative contribution of milk to the body Cs^{137} level varied from 54% in 1957 to only 23% in 1961 and that the average concentration of Cs^{137} in the balance of the diet increased from a minimum of 24 μc Cs^{137}/g K in 1957 to a maximum of about 50 μc/g K in 1960. In 1961, the concentration in the balance of the diet decreased, suggesting approximately a 1-year delay in peak level compare to milk.

The interrelationships among yearly average Cs^{137} fallout rate, concentration in milk, people, and the balance of the diet (excluding milk) are shown by the bar graph in Fig. 15. From these data and those

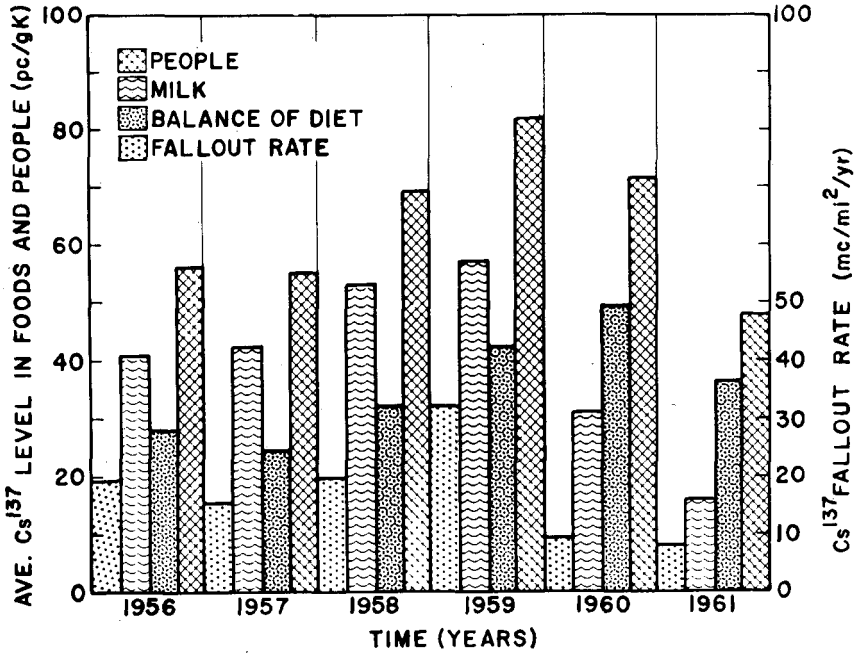


Figure 15. Yearly average Cs^{137} concentrations in people, milk, and balance of the diet and their relationship to yearly average fallout rate.

shown in Figs. 12 and 13, it may be concluded that, on the average, during the last 6 years the U.S. peak fallout rate occurred about mid-April, peak milk concentration occurred about mid-May, peak concentration in people occurred about the first of November, and peak concentration in the balance of the diet occurred about the first of April of the following year. Stated in terms of time lags, peak concentration in milk occurred about 1 month after the peak in fallout rate, the peak levels in people occurred about 7 months after the peak in fallout rate unless distorted by exceptionally large year-to-year changes, and the peak concentration in the balance of the diet occurred about 1 year after the peak fallout rate. An average production, distribution, and consumption time lag of about 1 year in the balance of the diet would therefore adequately explain the 7-month delay in the peak Cs^{137} concentrations in the population and why the levels in people (Fig. 13) did not start dropping during the weapon test moratorium until approximately 1 year after the precipitous drop in milk levels began. An average time lag of about 1 year between production and consumption of meats, canned goods, and frozen foods seems reasonable. The wide distribution of such foods also helps to explain the extremely small variation in body Cs^{137} levels throughout the U.S.

Radiation Exposure of the U.S. Population from Cesium^{137}

Radiation exposure from Cs^{137} fallout occurs both from material deposited on the ground and from that taken into the body. The external gamma radiation dose rate from Cs^{137} surface deposition may be estimated from the following expression:

$$\text{mr/yr.} = 5 \times 10^{-2} \times C \times E$$

where C is the Cs^{137} deposition level in mc/mi.^{2}, and E is the gamma ray energy (0.66 Mev). This expression gives the dose rate at about 3 ft. above an infinite plane surface in the open. It does not allow for the effect of shielding from buildings during time spent in-doors or removal of the Cs^{137} from the surface by leaching into the soil, weathering, or run-off. The United Nations Scientific Committee on Effects of Atomic Radiation (38) assumed that these phenomena would decrease the actual exposure to about 1/10th of the open field dose. The U.S. average Cs^{137} surface deposition levels as a function of time are shown in Fig. 16. These levels were derived from the Health and Safety Laboratory's Sr^{90} deposition data (37) by multiplying the Sr^{90} values by 1.7. The average Cs^{137} external radiation exposure of the U.S. population during the period 1952-1961 from all weapon tests prior to September 1, 1961, may be estimated from surface deposition data and the above equation. Integration of the area under the curve in Fig. 16 gives 498 mc yr./mi.^{2}, which, when substituted into the rate equation, gives a total open field exposure dose during the

period 1952-1961 of ~ 20 mr. When decreased by a factor of 10 to allow for shielding, etc., the average radiation exposure of the U.S. population from externally-deposited Cs^{137} is estimated at ~ 2 mr during the past 10-year period. Had weapon testing not been resumed, the 30-year genetic and 70-year bone marrow doses to people born in 1961 from externally-deposited Cs^{137} would have been ~8 and 13 mr, respectively. Preferential fallout in the North Temperate Zone latitudes of the fission products produced by the 1961 U.S.S.R. test series may result in a doubling of the Cs^{137} external genetic and bone marrow doses to the next generation of U.S. residents.

The average radiation exposure of the U.S. population from internally-deposited Cs^{137} may be determined from the population-weighted data given in Fig. 13. These data may be converted from pc Cs^{137}/g K to average adult body burden (in pc) by multiplying by 140 g, the average potassium content of a 70-kg standard man. The radiation dose rate from internal Cs^{137} may be estimated from the following expression derived from the Recommendations of the International Commission on Radiological Protection (39):

$$R = \frac{1.86 \times 10^7 \, f_2 \sum E(RBE)N \, q}{m}$$

$$1.57 \times 10^2 \, q$$

in which R is the dose rate in mr/yr., q is the Cs^{137} body burden in μc, m is the weight of the critical organ (in this case, 7×10^4 g),

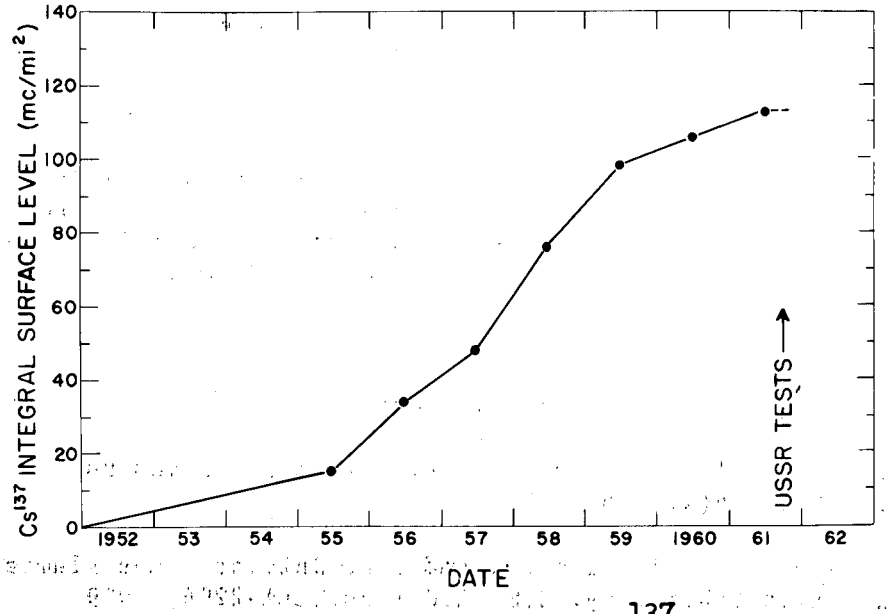


Figure 16. Trend in United States average Cs^{137} surface deposition level since beginning of thermonuclear weapon testing.

f_2 is the fraction of total body burden that is in the critical organ (1.0), and Σ E(RBE)N is an average energy term (Mev) weighted for the relative biological effectiveness and absorption of the radiation within the critical organ (0.59). Calculations on the basis of the above considerations and the integrated area under the population curve shown in Fig. 13 indicate that the average radiation dose to the U.S. population from internal Cs^{137} during the period 1956 through the third quarter of 1961 was ~8 mr. By assuming a linear rise from zero to approximately 45 pc Cs^{137} /g K from 1952-1955 inclusive and an exponential decline with a 10-month half-time beyond the third quarter of 1961, the total internal radiation exposure of the United States population from all weapon tests prior to the 1961 U.S.S.R. series would be ~11 mr, 2 mr prior to 1956, 8 mr during the period 1956-1961, and 1 mr after 1961. During the period of maximum dose (1959), when the average Cs^{137} level in the population reached a peak of about 82 pc/g K, the dose rate was 2 mr/yr. If instead of continuing to decline with a 10-month half-time the levels in the population were to have decreased with a much longer half-time of the order of 2 to 5 years, the added dose would be 1 to 4 mr.

As a result of spring 1962 fallout from the 1961 U.S.S.R. tests, the average Cs^{137} level in the U.S. population may increase from a 1961 low of ~ 35 pc/g K to a maximum of about 60 μc/g K in the last quarter of 1962. This will add very little to the integral radiation dose to the U.S. population. With the rather conclusive evidence that Cs^{137} levels in the population are largely related to fallout rate and its direct contamination of vegetation and not to the integral level in the soil, radiation exposure from internal Cs^{137} will be relatively significant only during periods of weapon testing. It is quite likely that external radiation from Cs^{137} deposited on the soil will contribute more to the 30-yr. genetic and 70-yr bone marrow doses than will internally-deposited material. In either case, radiation exposure of the U.S. population from past and present levels of Cs^{137} fallout seems to the authors relatively insignificant compared to normal variations in natural background rate or to existing guidance levels for radiation protection and does not seem to justify any large measure of concern.

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DISCUSSION

Dr. S. Allan Lough, Chairman of the session, directed the discussion of the papers of the session which follows.

LOUGH : Dr. LOUTIT made reference to studies in which I was interested, whether the discrimination factor in children was higher or not higher than had been observed in adults. We would like to make a similar study on a series of children of different ages to see how this discrimination factor changes with age. We thought we did pretty well to work on children 4 to 8 weeks old. We had a very well-controlled situation in a rather superbly operated orphanage in the Detroit area. If anyone can tell me how to get a 6 to 10 year old boy to collect all of his excreta, then we could proceed.

KNAPP : I should like to ask Dr. LOUTIT to comment on the points of view of, or difference of opinion between, Dr. LOUTIT and Dr. KULP on the discrimination factors for one-year-olds and those older than one year.

LOUTIT : I think the line was provided by Dr. COMAR in his presentation. Our monitoring program was initiated because of public concern. After having collected for 3 or 4 years, we didn't want to use the figures merely to allay public concern, but to see, what one could get out of their deduction.

We came to the conclusion that discrimination could be less in the earliest years of life than in the adult, but I don't believe this can be proved one way or the other by this sort of material. All it does is identify subjects for research.

Some, including our Chairman, have taken up the challenge and made experimental studies of a small number of cases, and found an O.R. in young infants 0.5 or greater. I understand Dr. STRAUB (U. S. Public Health Service, Cincinnati, Ohio) has

done similar work with similar results. Our own group took even younger infants, for very short periods it is true, and with Widdowson they have reported that about the age of one week after birth it might not even be possible to derive the O.R.; for instance, breast-fed infants had a positive Ca balance at that time but a negative Sr balance.

To sum up, all that results from monitoring can reliably do is to determine areas for research.

KULP : I would certainly agree with Dr. LOUTIT on this point. From the survey measurements alone one cannot determine whether there is a variation in the O.R. during the first year.

I was concerned primarily in showing that the model does fit empirically, which it does if you restrict the samples to the dietary area used in the calculation of the model.

I also wish to emphasize again, however, if the O.R. is as high as 0.5, then there must be a major compensating factor in the diet. This, of course, is not impossible. The HASL (Health and Safety Laboratory, U. S. A. E. C., New York) data indicate 14 $\mu\mu c\, Sr^{90}/g$ Ca for the diet of very young children when the adult average was 18 $\mu\mu c\, Sr^{90}/g$ Ca. This is not enough to account for the problem.

I would also like to point out that the data on the O.R.'s for one-year-olds are not yet published. This work is difficult experimentally. Until it is finally presented for critical analysis by other workers, judgment on its validity must be reserved. Going back 3 or 4 years we had meetings where competent experimentalists were getting anywhere from 2 to 5 for their discrimination factors in adults. Our work on human patients in cooperation with Dr. Laszlo at that time gave a discrimination factor of 4. A great deal more work needs to be done here. I think that ultimately the correct O.R.'s will be established in the laboratory. When it is, the total picture must be consistent with the monitoring data. You can't have a factor of two discrepancy between the model and the experimental data.

COMAR : Naturally, I would agree that the experimental approach in this particular instance would have to provide the answer. One has to be very careful with the survey results to take into account some of many factors that might change the situation.

For example, in very young children, there is a reservoir of bone laid down in utero that is subject to discrimination, across the placenta. Another point is that the mother might have used stable Ca pills under prescription during pregnancy.

If the children are breast-fed, this will lower the Sr/Ca value tremendously, by a factor of 10, breast milk having one-tenth the Sr^{90} that one would expect in the mother's diet. All such considerations will have to be taken into account.

Our experience in the past has been that whenever we have actually gotten the data on individuals (i.e., what they eat and what they retain) this has always come out in good agreement with predictions from experimental observations.

LOUTIT : I would like to volunteer something. In Dr. KULP's presentation, I understood him to say that the contamination of diet or milk in Wales was some 4 times that of the country at large, and this would bias our results. It is true that our children's bones did come largely from Wales, but the average milk value for the whole of that region is only 50% greater than the national mean. The values of 4 and 6 times the national or regional mean come from the very wet hill country, where dairy cows do not normally graze, though sheep do. I repeat, the milk supply does not come from the hills but from the lowland valleys, and has a mean value 50% greater only than the national mean. Furthermore, from Scotland, where an independent set of analysts are doing the bone work, the bone results are substantially the same as our combined United Kingdom results, where Wales is just a part of the national program.

KULP : There are individual areas in Wales that have 3 to 6 times the average milk level in the U. K. A large fraction of the bones plotted by Dr. LOUTIT were from Wales, thus it is not surprising that his points are much higher than our model curve which assumes a Sr^{90} concentration in diet similar to that in the average U. K.

For the year ending December 1959, in the U. K., the average value for milk (including Wales) was about 12 $\mu\mu c Sr^{90}/g$ Ca. The value for Wales was about 17$\mu\mu c$ Sr^{90}/g Ca. Therefore, if we were to take 1959, which was the particular year where we are discussing the problem, the difference between the ratio of Wales to the rest of England was close to a factor of 2. It would only have to be about 40% to account for the discrepancy on Dr. LOUTIT's plot.

Lora M. SHIELDS: (New Mexico Highlands University, Las Vegas, New Mexico): I should like to ask Dr. LANGHAM to what extent Cs^{137} enters the meat of grazing animals?

LANGHAM: Meat from grazing animals, for example beef and lamb, are especially high in Cs^{137}. In all probability, meats of this type provide another 25% of the body burden of Cs^{137} in man. In other words, the principal sources of Cs^{137} in man are, in the order of importance, milk, meat; and vegetables.

VAN MIDDLESWORTH: Do procedures such as double-label experiments show any difference in the Sr/Ca discrimination by one bone compared to another?

COMAR : As far as I know, on an over-all basis, there is very little difference in discrimination between different parts of the bone. Discrimination occurs primarily at the site of absorption in the gastro-intestinal tract, at the site of re-absorption in the tubules, at the placenta, and in the mammary gland. There seems to be only minor discrimination between the body fluids and the bones themselves.

KULP : In the work of Schulert and Laszlo, using Ca^{45} and Sr^{85} in terminal cancer patients, where they attempted to get the over-all discrimination factor, no difference in the ratio was found from one bone to another.

H. D. BRUNER : (Assistant Director for Medical and Health Research, Division of Biology and Medicine, U. S. A. E. C., Washington) I am sure we all regret that Dr. Laszlo is not here. The last time I saw him, he was still fighting the battle he waged unsuccessfully at meetings he attended here and elsewhere. I can't help but wonder whether the time is ripe, now that the experimental techniques are becoming sufficiently sophisticated for us to drop the O.R. and think of Sr in terms of its own metabolic units, as a separate member of the group of elements which are individually handled by the body. If it can vary independently of ionic Ca (and it does), why worry with the vagueness occasioned by O.R.'s and the attendant increased degrees of freedom?

COMAR : I have felt that there is a fundamental reason for considering Sr in terms of Ca namely, that the behavior of Sr is evidently governed by the level of the total alkaline earths, which includes both Ca and Sr. Since Ca predominates, it is the governing factor. I would cite a simple illustration. If an individual is eating 1g of calcium a day and, let us say, 20 $\mu\mu c Sr^{90}$/day, that individual will eventually develop in his body a value of 5 $\mu\mu c$ Sr^{90}/g Ca. If the same individual were eating twice the amount of Ca, but the same amount of Sr, he would develop half of the Sr body burden. What I am saying is: Here we have two situations in which the same amount of Sr is being taken in, but because of the Ca level of the diet, the body burdens differ by a factor of two. There are several other reasons but time does not permit discussion. I think this is really the fundamental one.

KNAPP : Some of the early attempts to estimate the amount of Sr^{90} that gets into people base their calculations on the amount of natural Sr that might be in soil and man. It was assumed that radioactive

Sr and natural Sr would act the same. That is the basis of the computation. It later was concluded that Sr^{90} followed Ca more closely than natural Sr. I have never understood whether or not there is some fundamental, logical difference between these two approaches, or whether there is a paradox involved here, or whether theoretically we have been able to reconcile the two approaches to computing Sr^{90} in bone from the amount on the ground. Is there anything worth mentioning that can be said on this subject?

LOUTIT : I think that I may sit on the fence by saying that, in the present decade, it is essential to consider Sr in relation to Ca. But I think that if we take the long view to the 1970's, it may then be more profitable to relate Sr^{90} to stable Sr. I think, certainly, we know enough at the present time to show that the idea of utilizing stable Ca as an isotope diluent of Sr^{90} will not work. In fact, it has been shown that adding stable strontium to a diet constant in Sr^{90} and Ca in fact increases the uptake of the Sr^{90} in some sort of mass effect.

I was wondering during Dr. KULP's presentation, whether the variations he described, the apparently high value in Australian children and the apparently high value in Copenhagen subjects compared with Bonn subjects and British subjects, might not be due to the high natural Sr that obtains in Australian soils and Australian vegetations? I regret I am ignorant of the stable Sr situation in Denmark. I don't know whether he has any observations on stable strontium for any of these localities.

KULP : No, we do not have any stable Sr measurements in foodstuffs from Australia. However, the stable Sr in the bones of Copenhagen people was not very much larger than the average for Western Europe. This still may be an important factor.

I would also like to comment on this from the point of view of the analytical problem. I think the reason we haven't used the specific activity of Sr in $\mu\mu c\ Sr^{90}/g$ Sr right from the beginning is that the Sr content of foodstuffs is poorly known. Part of this is due to analytical difficulty. You cannot analyze Sr reliably easily at low concentrations with wet chemical methods. You have got to do it by isotope dilution technique if you want good numbers. This is expensive. The number of laboratories that could do it are extremely limited. On the other hand, there are thousands of accurate Ca analyses on all foods, soils and bone from state surveys, federal surveys and other sources. I believe Dr. COMAR would agree that because we know the Ca concentration so well and can learn the discrimination factors that it is more useful at present to relate the Sr to Ca.

COMAR : We must decide what information is needed and then use the best approach. A brief comment on the points raised by Drs. KNAPP and LOUTIT: Even if we knew the Sr^{90}/stable Sr of all diets, this would only tell us the Sr^{90}/stable Sr to be developed in bone, whereas what we require to know is the Sr^{90} per unit mass of bone, or per amount of Ca since the Ca content of bone is relatively constant; it must be remembered that the amount of stable Sr per unit mass of bone is governed by the stable Sr/Ca of the diet.

I would take issue with Dr. LOUTIT about the effect of increasing stable Ca. Numerous long-term studies on reasonably normal diets indicate the expected inverse relationship between stable Ca intake and Sr^{90} body burden. Misleading results can arise, for example, from short term studies in which the individual has not adapted, and from abnormal diets in which phosphate levels are radically changed.

SUMMARY OF THE PROCEEDINGS

Staff of the Fallout Studies Branch
Division of Biology and Medicine
U. S. Atomic Energy Commission
Washington, D. C.

The many new findings which are contained in these proceedings cannot readily be reduced to a more condensed form than that in which they appear in the original papers. However, this summary is included on the assumptions that some readers, interested in the subject as a whole, will not find it convenient to study all the detailed material and that some of these as well as others may find useful a selection and appraisal of the highlights of the Conference. It should be borne in mind in reading the following that in addition to the speakers whose names are mentioned, their co-authors and their references, many of the results can be further supported by the work of others, or are based on sample collection and analysis programs partially or wholly sponsored by other Government agencies.

Local Fallout

Studies of physical, chemical and radiological characteristics of fallout at early times after detonation have had to rely mainly on such experimental data as were obtained prior to the test moratorium. Qualitatively it is well established that the different radionuclides produced in a nuclear detonation are fractionated, that is their ratios to one another differ in different particle size fractions, in different radial shells within each particle and in different parts of the "stabilized" cloud. For example, Sr^{90}, formed mainly by radioactive decay from a short-lived krypton isotope, is enriched in the particles which appear in the cloud later and are therefore smaller, and is depleted in the larger particles as compared with a fission product such as Zr^{95} which does not have a volatile precursor. Sr^{90} is also enriched in the outer layer of accreted material and depleted in the central core of the particles which have solidified from liquid or vapor. Consequently,

Zr^{95}/Sr^{90} ratios in fallout vary with time after detonation and, therefore, with distance downwind. Even the ratio Cs^{137}/Sr^{90} shows pronounced fractionation in local fallout (LARSON), both having volatile precursors but of different half-lives. This fractionation is most pronounced in the case of low-yield, land surface bursts and least in the case of air bursts and very high yield bursts.

The particle size distribution in the debris cloud from a land surface burst and in the fallout shifts with time towards the smaller sizes which contain a smaller proportion of earth material. Thus the fraction of each nuclide which is soluble or biologically available tends to increase with time and distance from the point of burst. The gamma energy spectrum and decay rate of the fallout also must vary with time and distance as well as with conditions of burst.

General quantitative models are being developed (FREILING) to predict these phenomena as a function of energy yield, chemical composition of weapon and environment and height of burst. Many of the more striking fractionation phenomena are accounted for by these models, but many difficult problems remain. These include lack of adequate basic data on fireball cooling processes and on condensation of complex mixtures of vaporized materials; irregularity and obscurity of turbulent mixing processes and flow patterns within the cloud; and gross departure of particle trajectories and fall times from those attributable to gravitational sedimentation from a "stabilized cloud" altitude (presumably due to atmospheric eddy motions at early times and largely to precipitation scavenging at greater times and distances).

Experimental data bearing on these physical-chemical problems are extremely scanty (LARSON). Measurements at the Nevada Test Site show a low ratio of biologically available to total Sr^{90} within a few miles from the point of burst as compared with greater distances, also a low ratio of Sr^{90} to total fission products. These are in the direction expected and quantitatively very substantial. A further observation suggesting reduced entry of radionuclides into the food chain in areas of local fallout from surface bursts is that of selective shedding of the larger particles by larger particles by leaves. On the other hand, the high proportion of very small particles coming to the ground at short distances downwind from both tower and balloon shots has not yet been explained.

Irregularities and "hot spots" have been noted in the distribution of fallout within the first few hundred miles. Irregularities of terrain are one suspected cause (LARSON). Small-scale irregularities of the wind field in time and space are also large enough to suggest that these may also be an important cause (ALLEN). Closely spaced upper wind observations in space and time have permitted substantial improvement in predicting the direction of the major axis ("hot line") of the fallout pattern over what can be done with conventional meteorological observation networks, but the meteorological data are still inadequate to

account for the smaller scale structure of the patterns.

Even with the considerable range of yields and conditions of bursts which have been experienced in nuclear tests conducted to date, conditions of interest in military and civil defense planning have not been simulated sufficiently closely, nor have sufficiently complete measurements of local fallout been made at the higher yields, to give much confidence in the existing fallout prediction models for these applications (FERBER). Analyses of the available data by different groups for different purposes have resulted in models which for given burst conditions in the MT range, predict distances for a given dose contour varying by a factor of about 3 to 4, total area integrals of dose within a given contour varying by a similar factor and dose rates at a given location 1 hr. after detonation varying by an order of magnitude. Predicted fatalities under given conditions of shielding can vary even more widely. The variations result largely from uncertainties regarding the scaling laws for vertical distribution and particle size distribution of radioactivity in the stablized debris cloud. Additional sources of variation are introduced by incorporation or neglecting variations of wind shear and by accepting various fractionation, gamma energy and decay laws for the deposited debris.

It appears that some improvement can be made in the prediction models for local fallout by more organized studies bringing to bear on the problem the large and complex body of available theory and experimental data. However, the phenomena include some such as high-temperature condensation thermodynamics of mixed vapors, cloud dynamics and turbulent mixing, for example, for which no satisfactory theory exists. It is possible that much of the uncertainty will remain unless suitable measurements are obtained in connection with actual nuclear detonations over a range of conditions.

Tropospheric Fallout

That portion of the debris cloud which stabilizes in the troposphere and which is not deposited in the first 12 to 24 hrs. consists of sufficiently small particles that, at least under weather conditions such as those which have generally been selected for conducting weapons tests, the levels of fallout are largely determined by the atmospheric vertical motions and precipitation processes occurring along the trajectory of the cloud. Since the total amount of fission debris stabilizing in the troposphere from a given detonation is limited (only low-yield clouds remain entirely in the troposphere; with increasing yield above a few hundred KT, the fraction remaining in the troposphere decreases to a very small percentage), and since the persistence of the tropospheric debris is relatively brief (on the order of a month) but sufficient to permit widespread dispersal, it has not been possible to carry on any new studies of tropospheric fallout during the test moratorium. Both the accumulated

and the incremental contamination of the environment during this period have been dominated by stratospheric fallout.

Stratospheric Fallout

The discovery of a natural population of hygroscopic sulfate particles about 0.03 to 1.0 μ in radius, in an apparently world-wide layer of nearly uniform character between 16 and 24 km in altitude (MANSON), has important implications. It appears from size-spectrum considerations that the rates of formation and coagulation are such that the sedimentation rate may play a significant part in determining the residence time of any smaller particles introduced in this layer. A positive correlation between sulfur content and total β activity in aircraft impactor samples of particulates (about 0.4 μ) collected at about 20 km supports the tentative conclusion that the radioactive debris particles go along with the sulfur in this process. However, comparison with filter samples which are believed to collect essentially all particles suggests that the debris particles may not coagulate beyond about 0.01 μ and in particles above this size possibly become distributed uniformly with particle number rather than particle mass. This might limit the importance of sedimentation as a mode of stratosphere-troposphere transfer even in the lower stratosphere, where the high tropopause in relation to the height of the sulfate aerosol layer and the absence of vigorous atmospheric mixing processes would tend to favor it.

At higher (polar) latitudes the bulk air-exchange processes probably dominate over the relatively slower precipitiation by sedimentation of sulfate particles. At higher altitudes approaching 30 km where samples have been collected by balloon-borne impactors (MARTELL), the size distribution of the radioactive debris seems to shift strongly toward smaller radii. At 20 km most of the radioactivity has been found in the size range between a few hundredths and a few tenths μ radius. In the few samples collected near 30 km, both in Minnesota and in India, most of the radioactivity was found in the size fraction less than 0.02 μ.

The distribution of both artificial and natural radioactivity in the stratosphere and the time-scales and mechanisms of transport and deposition by various processes have received considerable study and a number of significant advances have been made in understanding these phenomena.

Stratospheric and tropospheric inventories of Sr^{90} have been calculated at 6-month intervals based on data obtained in the balloon, aircraft and ground-level air sampling programs (MACHTA). When added to the inventory on the surface of the earth obtained from pot, funnel and soil data, the total agrees reasonably well with the total deduced from available information on yields of past tests.

By using W^{185} as a tracer for 1958 equatorial tests and Sr^{89} as a tracer for all 1958 tests it has been possible to resolve the Sr^{90} fallout as measured by the world-wide pots and funnel network into several components (MACHTA). Sr^{90} fallout from the fall 1958 U.S.S.R test series, conducted about 70° N, was nearly all confined to the Northern Hemisphere, was peaked in middle latitudes, and was virtually complete by the summer of 1959. That from the U. S. HARDTACK I equatorial test series in 1958, also peaked in middle latitudes, was distributed in both Northern and Southern Hemispheres with most in the Northern.

The Sr^{90} fallout during early 1960, apparently consisting largely of equatorial test debris from HARDTACK and earlier series, was distributed similarly to the 1958 W^{185} fallout. During late 1960 and early 1961, however, the Sr^{90} fallout in the two hemispheres was nearly equal. During this time a significant contribution was being made by fallout from the 1958 high altitude detonations as evidenced by the tropospheric levels of Rh^{102}, a tracer produced in the ORANGE shot (GUSTAFSON, KREY).

Both surface air concentration and deposition measurements have now confirmed that a spring maximum and autumn minimum of stratospheric fallout occur even in the absence of recent test activity, since this seasonal cycle was quite pronounced in the Northern Hemisphere and reasonably well established in the Southern Hemisphere during both 1960 and 1961 (MACHTA, LOCKHART, GUSTAFSON, KURODA).

It is also clear that tropospheric concentrations and deposition rates of stratospheric debris are higher in middle latitudes of both hemispheres than near the equator regardless of the latitude or altitude of stratospheric injection (MACHTA, LOCKHART). It has been difficult to establish the trend of concentration in air or precipitation from middle latitudes poleward although total deposition decreases with decreasing precipitation.

The resolution of sources has also led to the following estimates of residence times for debris introduced at different locations in the stratosphere (MACHTA, GUSTAFSON). Most of the debris from the U.S.S.R. Arctic tests of 1958 was deposited the first spring season after injection. Debris from equatorial tests in the low MT range exhibited a half residence time of the order of a year or less during the first year, increasing somewhat in later years. Debris injected at a very high altitude (exceeding 40 km) near the equator began to appear at low altitudes about a year after injection, and then exhibited a deposition rate corresponding to a residence time of at least 5 yrs.

Early estimates of mean stratospheric residence times of the order of 5 to 10 yrs. based on measurements of fallout from equatorial debris clouds (from the higher yield detonations of Operations IVY and CASTLE) which may have reached altitudes of 30-40 km appear to fit into this pattern.

It is not known what part of the holdup of high altitude equatorial debris is due to a slow rate of horizontal transport to higher latitudes, what part is due to slow downward transport and how these might vary with

altitude and latitude of injection. In particular, it is no known whether the world-wide fallout pattern from a debris cloud such as that introduced by the U.S.S.R. 55-60 MT detonation in October 1961, at an estimated altitude of 40-50 km near the north pole, will more closely resemble that of the equatorial high altitude debris or that of the Arctic low stratosphere debris.

In attempting to account for geographical and seasonal non-uniformities of stratospheric fallout any or all of the following processes could be involved:

(1) circulation or mixing processes within the stratosphere;
(2) exchange processes between the stratosphere and troposphere;
(3) circulations or mixing processes within the troposphere and
(4) deposition processes.

Each of these categories has received some study and each apparently makes an observable contribution to the observed pattern.

In the stratosphere (BARNES, NEWELL) two kinds of processes seem to be important. It appears that mixing by large-scale turbulent eddies along surfaces of constant entropy which slope downward from the equator toward the poles may be adequate to account for the transport of W^{185}, as well as ozone, heat and momentum, during the winter and spring. Evidence for a systematic poleward circulation is ambiguous. Systematic circulations may, however, play a dominant role in transporting ozone equatorward during the summer and fall. Internal heating and cooling within the stratosphere appears to be a significant driving mechanism for this latter circulation. While these findings suggest ways in which the stratospheric motions might lead to a late winter or spring maximum of the stratospheric debris concentration at high latitudes they are still far from conclusive.

Processes which would influence transfer from the stratosphere to the troposphere do appear to have a strong seasonal and latitudinal variation (STALEY, DANIELSEN). These processes include the spring upward migration of the tropopause as well as the frequency and intensity of tropospheric weather disturbances likely to involve incursions of stratospheric air. This latter concept pictures the "tropopause gap" as the statistical average of sporadic pulses.

Deposition of Sr^{90} and other products of weapons tests, either per unit time and area or per unit volume of precipitation, show large variability even after the stratospheric distribution has become relatively uniform (DINGLE, KRUGER, KURODA). In addition to large seasonal variations, there are substantial variations from one storm to another and within the course of a single storm. Effects of the duration of exposure of the air mass to precipitation cleansing, concentration of raindrop contaminants by evaporation in falling through unsaturated air, and penetration of precipitating

cloud systems into the tropopause region or into upper tropospheric air of relatively recent stratospheric origin have been rather convincingly indicated (KRUGER). The last mentioned may be a systematic phenomenon in squalls and showers of the type which characterize the middle-latitude spring season (DANIELSEN).

Despite these variations in concentration, however, ratios of concentrations of different radionuclides in precipitation exhibit remarkable stability. Not only the values of such ratios but even their rates of decay during a succession of rain storms can be used to characterize the sources contributing to the fallout (KURODA, KREY, COLLINS). Another encouraging evidence of consistency is the fact that gamma radiation dose rates due to cumulative levels of fallout of long and intermediate-lived fission products over peiods of many months, calculated from gamma ray spectrum analysis of soils at Chicago (GUSTAFSON), agree with similar data obtained by radiochemical analysis of precipitation samples collected at New York (COLLINS).

Deposition and Movement in Soils and Ground Water

A study was made to obtain relationships between Sr^{90} deposition and rainfall rates in Clallam County, Washington (ALEXANDER). This limited geographical area was selected for detailed study because of the marked variation of annual amounts of rainfall from site to site. This study showed, as has been indicated by other studies, that Sr^{90} deposition varies with the amount of rainfall. However, it was concluded that there is some deposition as dry fallout. In this area it was estimated that 16 m_c $Sr^{90}/mi.^2$ was deposited as dry fallout up to the fall of 1960. Total deposition at the 5 sites ranged from 34 to 137 mc $Sr^{90}/mi.^2$.

Results of soil sampling at a large number of sites throughout the world in 1960 indicated low total Sr^{90} deposition on the average between 1959 and 1960. The global average increment during the year appears to be less than 5 $mc/mi.^2$ which may be less than the analytical error associated with a single result. Because of this fact, annual increments calculated for a single site from 1959 and 1960 soil collections and analyses are unreliable. The much better estimates based on pot and funnel fallout collections confirm the small magnitude of the increment. Evaluation of the 1960 results is currently in progress.

Studies performed by the Soils Laboratory of the U. S. Department of Agriculture concerning runoff of Sr^{90} (MENZEL) included small watersheds, small plots and river water. From small watershed studies it appeared that loss of Sr^{90} was probably due to surface runoff. The amount lost by crop removal probably did not exceed 5% of the Sr^{90} fallout while leaching loss probably did not amount to more than 15% of accumulated fallout. Since somewhat lower Sr^{90} contents of soil were found at lower contours,

an effect of length of slope was suggested. There was little or no accumulation of Sr^{90} in the area of silt deposition so that Sr^{90} must have been carried off the watersheds. In runoff studies from small plots, loss of Sr^{90} was shown to be affected by vegetative cover and the amount of soil eroded from the plots. Plots having good vegetative cover showed the least loss of Sr^{90}, soil and water. Crop cover appeared to have little effect on the amount of Sr^{90} transported per inch of water runoff, however. Analyses of river water and precipitation in the Ohio River basin by the Public Health Service have indicated that from 4 to 12% of the Sr^{90} in precipitation appeared in the river water. Amounts of Sr^{90} appearing in river waters are consistent with results obtained from watershed and small plot studies of runoff. The amount of Sr^{90} deposited in impounded waters is still largely unknown, however.

Ground-water studies (CLEBSCH) conducted by AEC and USGS have shown that very little fallout radioactivity enters ground water after is is deposited on soil surfaces. In general sedimentation and natural filtration by soils and rock are quite effective in preventing appreciable contamination of ground water. Since fallout material consists of much particulate matter it seems reasonable that a great deal of the radioactivity is retained in top layers of soil. This has been indicated also from studies of concentration vs. depth of soil studies. Soluble materials, such as Sr^{90} are effectively absorbed by soil particles or the various rock formations. Other important factors are dilution in large water reservoirs and the very slow rates of movement of water from the surface into ground-waters as well as the very slow movement of water within a ground-water reservoir. Hold-up times due to this slow movement and the filtration and low-solubility factors are sufficient for appreciable decay to occur. The effectiveness of these processes in preventing heavy fallout contamination of ground-water varies with depth of reservoirs or water table. In the event of very heavy fallout as from a nuclear war, ground-water contamination may become more important, although surface water supplies would present much greater problems. Fallout tritium has been of interest since it provides a tracer for water itself and has been used to study ground-water movement and mixing as well as other hydrologic phenomena.

Cycling of Fallout Nuclides in the Biosphere

Of further importance in the consideration of fallout and its effects on man is the distribution in biological populations in the environment and the part played by biological systems. From the standpoints of external radiation and entry of radionuclides into the food chain, the distribution and environmental cycling of fallout radionuclides has received increasing attention. Since research results pertaining to these aspects of fallout radionuclides were reviewed in considerable detail at the First National Symposium on Radioecology at Fort Collins, Colorado, in September 1961, sessions of that symposium which concerned

cycling of fallout nuclides in the terrestrial, freshwater and marine environments were summarized at the present conference (AUERBACH, SEYMOUR, KRUMHOLZ). Since these summaries in themselves cover research findings of a very large number of investigations, no further reduction of these is attempted here. The papers and discussion include such topics as concentration and transport by insects, birds and aquatic organisms, uptake by marsh and pond plants, and effects of grass burning on contamination of subsequent growth.

Fallout Nuclides in the Food Chain and Man

Studies of stable Sr and stable Ba in human bone and diet in Canada (GRUMMITT) indicated a total daily intake of Sr of 1.29g and of Ba 0.40g. Ba was found to be much more variable in individual food items than was Sr. While milk is the only major contributor of Sr to the Canadian diet, Ba appears to be derived about 25% from milk, 25% from flour, and up to 25% from potatoes. Stable Sr in the skeleton was found to be distributed nearly normally; there was no apparent tendency to concentrate in a particular bone. The population distribution was quite skewed toward the high side, however; quite different from results reported for New York City. There appeared to be no age effect in this study since averages for 0 to 19 and 20 to 70 yr. age groups were 300 and 320 μg/g Ca respectively. The population distribution for stable Ba was similarly skewed with values up to 5 times the mean, and more scatter was observed for Ba than for Sr. More scatter was apparent within the skeleton also. The Ba and Sr results showed no correlation, indicating a dietary control rather than a metabolic one. Sr/Ca O.R.'s (bone/diet) were found to be about 0.25 with the mean about 15% higher than the mode. The mean of Ba/Ca O.R.'s (bone/diet) was 0.07 and the mode was 0.05. The Ba/Sr O.R.'s had a mean of 0.28 and a mode of 0.25, quite similar to those for the Sr/Ca O.R.'s.

From the tri-city diet studies at HASL and a review of other data prediction models for Sr^{90} in the diet were presented (RIVERA) and estimates were made of dietary concentration of Sr^{90} in New York City and San Francisco resulting from the recent U.S.S.R. nuclear test series. The prediction model provides for fallout rate and cumulative deposition factors. This model is believed to be good within a factor of 2 or 3. Models for the various major dietary components were discussed. KNAPP discussed his previously reported model for predicting monthly U. S. average levels of Sr^{90} in milk. Although the available models are, of course, still based on a number of assumptions, it appears that considerable progress has been made in developing methods for estimating dietary levels of Sr^{90}/Ca where nuclear yield or, better still, deposition data are available.

A status review was presented (COMAR) on selected aspects of food surveys, as well as on strontium, cesium and iodine as related to fallout problems.

It is clear that food surveys should be designed in relation to the required objectives. Results to date suggest that vigorous sampling procedures may not be justified for many practical purposes. OR body/diet $\left(\text{observed ratio } \frac{\text{Sr/Ca of body}}{\text{Sr/Ca of diet}}\right)$ values for adult man range from 0.16 to 0.5, with values based on most extensive sampling falling near 0.25. Values are probably higher, 0.5 and above, for the very young; the significance of the higher values in respect to harm cannot yet be assessed. ^{OR}milk/diet values for man, cow and goat are about 0.1. On typical rations the concentration of fallout Sr^{90} in milk was halved by doubling the calcium content of the diet in a long-term study. This confirms predictions from short-term experimental studies. Secretion of radioiodine into milk is highly variable. Typically the dairy cow secreted about 1% of the ingested daily intake per liter of milk. Attention is called to the differing abilities of various species to concentrate iodine from blood plasma to milk. About 8% of the daily radioiodine intake is found in the egg of the laying hen, with most being found in the yolk and a significant proportion bound to protein. After a single administration the yolk does not reach a maximum until about the sixth day. The turnover time of cesium in various species was reviewed, and the need stressed for consideration in terms of multi-component exponential behavior. In monogastric species the overall turnover time appears to correlate with body size, ranging from a few days for the mouse to about 100 days for man. In ruminants the values are relatively smaller, being a few days for the goat and somewhat longer for the cow. Special note was made of the possibility that small amounts of radiocesium may be trapped in the skeleton and thus not be available for exponential removal. In the dairy cow, about 1.5% of the daily ingested radiocesium was found per liter of milk, or about ten times the corresponding figure for radiostrontium.

Preliminary results were presented (BIRD) of a study of milk sampling for Sr^{90} content in Canada. From preliminary findings in a limited number of areas there was no statistically significant difference in mean Sr^{90} estimates for milk between "grazing" classes, nor between days of sampling within 5 consecutive days for the same town. There was a significant variation between towns. Morning readings were higher than afternoon readings but there was no significant difference between the means of the two periods of sampling. Because of the preliminary results reported here and the seasonal and geographic variations interpretations of results must be made with caution. Further investigation of sampling design is indicated.

A review of human bone surveys for Sr^{90} and stable Sr and Ca in the United Kingdom (LOUTIT) provided several interesting results. Some of these are similar to results reported of studies in the U. S., although both studies are only briefly summarized here. From these studies in the U. K. the O. R. (fetal bone/maternal plasma) was 0.625. The child at birth is capable of discrimination, probably mainly by the renal

mechanism, and it improves this capability with age. The O. R. (bone/diet) falls from about 0.5 to about 0.25 or below, largely due to the gastro-intestinal process. The rate of turnover of pre-existing bone salt can be estimated in infants by consideration of the relative specific activities (Sr^{90}/Ca) of diet and bone. It is concluded that skeleton turnover must be nearly 100% in the first year of life and very high in the second. Only with reliable data on infants' diets obtained from surveys could representative figures be produced. Based on specific activities (Sr^{90}/Sr) of the bones and the estimated diets it is estimated that the skeletal turnover is about 10%/yr. in the third to eighth years, after which it falls to very low values during the second burst of growth around puberty. In adults, by calculations from specific activities (Sr^{90}/Sr) of bones and the national diet it is estimated that mainly ivory bones such as shaft of femur, tibia and calvarium of skull turn over about 1% of their mineral per year; the highest mean annual rate of turnover was in vertebrae, 8%.

KULP summarized his analysis of data accumulated over several years with respect to Sr^{90} in human bone. Reasonably good predictions can be made of Sr^{90}/Ca levels in the world population for a given input of atmospheric contamination, based on surveys following past nuclear test series. The maximum concentration in fetuses in the Western culture Temperate Zone was reached in 1959 at about 1.2 $\mu\mu c$ Sr^{90}/g Ca and decreased rapidly in 1960 and 1961. Fetal bone ratios of Sr^{90}/Ca were estimated to be about 8% of that in the adult diet. The turnover rate of Sr and Ca in adult bone appeared to be about 2%/yr., although this rate appears to vary with individual bones of the skeleton. Although the total deposition in the Southern Hemisphere has been only 1/4 that of the Northern Hemisphere, bone levels in the Southern were found to be about 1/2 those of the Western culture populations in the Northern Hemisphere, probably attributable in part to dietary differences.

A final summary was reported (LANGHAM) of the LASL study of Cs^{137} in milk and people in the U. S. which was completed in 1961. The study beginning in 1956 included Cs^{137} and K^{40} determinations in powdered milk samples collected at 55 sites in the U. S. and Canada, and in people visiting LASL who volunteered to be counted. In the analysis of data consideration was given to agricultural conditions, dairy practices, meteorological conditions, milk production and geographical location. Time-trends in U. S. powdered milk showed regular seasonal fluctuations at all stations with maxima generally occurring in spring and minima in late summer, following a similar pattern to fallout rate with alterations due to feeding practices. Tropospheric fallout from Nevada tests in 1957 and 1958 seemed to cause maxima later in the year. A large number of factors may influence these trends. The rapid response of Cs^{137} levels in milk to change in fallout rate strongly suggests predominance of rate over cumulative deposition, and direct contamination of vegetation rather than uptake through roots is suggested as a predominant mechanism. Using Sr^{90} fallout data from the Midwest for 1957-1961, for which adequate data were available,

and testing several empirical relationships between fallout rate and milk levels of Cs^{137}, a formula was obtained relating time-weighted fallout rate and milk levels. It was observed from the results of this analysis that there was little or no evidence of uptake of Cs^{137} from soil. When considering geographical distribution milk levels appeared to be influenced primarily by factors other than integral surface deposition. Attempts to deduce quantitative relationships between annual precipitation and yearly average milk levels of Cs^{137} were not very successful, only crude correlations were obtained.

Time-trends in people from New Mexico show seasonal fluctuations as do fallout rate and milk levels of Cs^{137}, although the pattern in later years is 6 to 8 months out of phase with those prior to 1959. Average agronomic, metabolic and food distribution and marketing time lags are important factors here. Beginning in 1959 perturbations in the population level of Cs^{137} occurred which may be due, in part at least, to testing schedule, yield and location of tests in 1958. From an early 1960 maximum the population Cs^{137} levels dropped to a new minimum in mid-summer, possibly reflecting increased stratospheric fallout rate and feeding of stored feed early in the year. The data strongly indicate a change in pattern of maxima and minima in Cs^{137} levels in people following cessation of nuclear tests. From the peak value in 1956 Cs^{137} levels in the New Mexico people dropped with an apparent half-time of about 9 months, from the 1960 peak about 12 months and during 1961 about 8 months; or an average of about 10 months.

When geographical regions were considered with respect to 6-yr. average Cs^{137} levels in people the outstanding feature was the very small variation over the U. S. Averages for 10 regions (not population averages) ranged from 46 to 71 $\mu\mu c$ Cs^{137}/g K. Geographical distribution data of others were in good agreement with the LASL study. These results may be due to sampling bias, and in part undoubtedly to the wide food distribution in the U. S. Relative differences in geographical areas were found to be consistent for each year of study. Frequency distributions for all U. S. adults measured showed deviation from normal for all years except 1956 and 1961, and was especially pronounced in 1959. For 1959 the data appeared to fit a log-normal distribution better but still showed an upward deviation on the high level side. From analysis of inter-relationships over the last 6 years it appeared to fit a log-normal distribution better but still showed an upward deviation on the high level side. From analysis of inter-relationships over the last 6 years it appeared that in terms of time lags, peak concentration in milk followed the peak fallout rate by about 1 month, that in the balance of the diet by about 1 yr. and that in people by about 7 months.

Summary Comments

The research on fallout during the three-year test moratorium has quite naturally made greatest advances in those areas related to long-lived radionuclides such as Sr^{90} and Cs^{137} which have continued to come down from the stratosphere. Other areas have progressed relatively slowly. These include many aspects of local fallout from surface bursts and the systematic and random variations of environmental levels and body burdens of short-lived radionuclides in tropospheric fallout. Attempts to determine transfer coefficients and time lags from one step to the next in the fallout cycle still suffer from dependence on survey data obtained from relatively poorly coordinated sampling networks, although the situation is improving considerably in this regard. On the whole, there has been an impressive sharpening of the tools for predicting and assessing various modes of human radiation exposure due to fallout from a wide variety of sources; still, the available tools are not so keen as to give cause for any relaxation of effort.

Registered Participants

The 254 registered participants in the conference are listed below.

Harold H. Abee, ORNL, Oak Ridge, Tenn
Philip H. Abelson, Geophysical Lab., Wash., D. C.
Robert B. Adams, Western Reserve Univ., Cleveland, O.
Lyle T. Alexander, USDA, Beltsville, Md.
James S. Allen, USDA, Beltsville, Md.
Philip W. Allen, U. S. Weather Bureau, L s Vegas, Nevada
Robert E. Allen, AEC, Wash., D. C.
John Ambler, USDA, Beltsville, Md.
Calvin E. Anderson, U. S. Weather Bureau, Wash., D. C.
Frederic D. Anderson, AEC, Wash., D. C.
George T. Anton, AEC, Wash., D. C.
Frank J. Arsenault, AEC, Wash., D. C.
Stanley I. Auerbach, ORNL, Oak Ridge, Tenn.
John G. Bailey, USPHS, Wash., D. C.
Major David T. Baker, Hq. Dept. of the Army, Wash., D. C.
Robert E. Baker, AEC, Wash., D. C.
Arnold A. Barnes, J., Mass. Inst. of Tech., Cambridge, Mass.
Lester A. Barrer, N. J. State Dept. Health, West Orange, N. J.
Stan Batten, RAND Corp., Santa Monica, Calif.
Lewis Battist, Nuclear Science and Engrg. Corp., Pittsburgh, Penn.
E. J. Bauser, Joint Comm. on Atomic Energy, U. S. Congress, Wash., D.C.
Christian Beck, AEC, Wash., D. C.
Harold Bernard, AEC, Wash., D. C.
Charles E. Billings, Harvard Sch. Public Health, Boston, Mass.
P. M. Bird, Dept. National Health and Welfare, Ottawa, Canada
A. Bertrand Brill, USPHS, Wash., D. C.
Robert W. Black, Naval Research Lab., Wash., D. C.
Ernest W. Bloone, Nuclear Defense Lab., Army Chem. Center, Md.
Lester E. Boleyn, USDA, Beltsville, Md.
Bruce Bollermann, Altantic Research Corp., Alexandria, Va.
Robert G. Bostrom, USPHS, Wash., D. C.
E. H. Bouton, Nuclear Defense Labs., Army Chem. Center, Md.
Vaughn T. Bowen, Woods Hole Oceanographic Inst., Woods Hole, Mass.
S. S. Brar, ANL, Argonne, Ill.

H. David Bruner, AEC, Wash., D. C.
James O. Buchanan, Off. Emergency Planning, Wash., D. C.
William W. Burr, AEC, Wash., D. C.
Ernest D. Campbell, AEC, SFOO, Berkeley, Calif.
Salvador N. Ceja, AEC, Wash., D. C.
Alfred Clebsch, Jr., U. S. Geol. Survey, Wash., D. C.
Roger J. Cloutier, ORINS, Oak Ridge, Tenn.
Ronald F. Coene, USPHS, Wash., D. C.
Stanton H. Cohn, BNL, Upton, L. I., N. Y.
William R. Collins, Jr., AEC, HASL, N. Y.,N. Y.
Cyril L. Comar, Cornell Univ., Ithaca, N. Y.
Walter C. Conover, U. S. Army Signal Research and Development Lab., Ft. Monmouth, N. J. (SIG-RL/SMA)
Enrico F. Conti, AEC, Chicago Oper. Off., Argonne, Ill.
Capt. Burton J. Conway, Nuclear Defense Lab., Army Chem. Center, Md.
D. John Coparanis, U. S. Weather Bureau, Wash., D. C.
Capt. F. Costagliva, Military Liaison Comm. (AEC), Wash., D. C.
David Costello, AEC, Wash., D. C.
Frederick P. Cowan, BNL, Upton, L.I., N. Y.
Russell F. Cowing, New England Deaconess Hospital, Boston, Mass.
Raymond G. Cragle, Univ. Tenn. - AEC, Agricult. Research Lab., Oak Ridge, Tenn.
Harold W. Crouch, Eastman Kodak Co., Rochester, N. Y.
Donald V. Crowe, General Mills, Inc., Minneapolis, Minn.
Edwin F. Danielsen, Univ. of Washington, Seattle, Wash.
Arve H. Dahl, USPHS, Wash., D. C.
Robert L. Darneal, AEC, Wash., D. C.
Robert A. da Roza, Univ. of Calif. LRL, Livermore, Calif.
A. K. Davis, USPHS, Wash., D. C.
Jared J. Davis, Hanford Labs., Richland, Wash.
L. A. Dean, USDA, Beltsville, Md.
Robert F. Dever, USDA, Beltsville, Md.
D. A. Dietsche, General Mills Inc., Minneapolis, Minn.
A. Nelson Dingle, Univ. Michigan, Ann Arbor, Mich.
Capt. J. A. O'Donoghue, Defense Atomic Support Agency, Wash., D. C.
Peter J. Drevinsky, Air Force Cambridge Research Labs., Bedford, Mass.
Charles L. Dunham, AEC, Wash., D. C.
Gordon M. Dunning, AEC, Wash., D. C.
W. H. Durum, U. S. Geological Survey, Wash., D. C.
John A. Eckert, USPHS, Wash., D. C.
Abraham Edelman, Nuclear Science & Engrg. Corp., Pittsburgh, Penna.
Raymond R. Edwards, Nuclear Science & Engrg. Corp., Pittsburgh, Penna.
Surg. Capt. F. P. Ellis, RN, British Naval Staff, Main Navy Bldg., Wash., D. C.
Carl A. Erickson, The Boeing Co., Seattle, Wash.
Lee E. Farr, BNL, Upton, BNL, Upton, L. I., N. Y.
Thomas R. Fewell, USPHS, Wash., D. C.
Frederick G. Finger, U. S. Weather Bureau, Wash., D. C.
Birney R. Fish, ORNL, Oak Ridge, Tenn.

Edward C. Freiling, U. S. Naval Radiological Defense Lab., San Francisco, Calif.
Maurice H. Frere, USDA, Beltsville, Md.
Sheldon F. Friedlander, John Hopkins Univ., Baltimore, Md.
James P. Friend, Isotopes Inc., Westwood, N. J.
Charles R. Fordyce, Eastman Kodak Co., Rochester, N. Y.
Eric B. Fowler, LASL, Los Alamos, N. Mex.
James J. Fuquay, Hanford Labs., Richland, Wash.
James A. Garrett, Jr., AEC, Wash., D. C.
Robert G. Gast, Univ. Tenn. - AEC Agricultural Research Labs., Oak Ridge, Tenn.
K. C. Giles, U. S. Weather Bureau, Wash., D. C.
Maj. Jack K. Gilham, U. S. Air Force (AFCIN-3K2), Arlington, Va.
H. Bentley Glass, Johns Hopkins Univ., Baltimore, Md.
Jack C. Greene, Off. Emergency Planning, Wash., D. C.
William E. Grummitt, Atomic Energy of Canada Ltd., Chalk River, Ont.
Phillip F. Gustafson, ANL, Argonne, Ill.
F. Haghiri, Ohio Agr. Expt. Station, Wooster, O.
Dave Hall, Health Physics Off., Army Chem. Center, Md.
Luther M. Hardin, Nuclear Defense Lab., Army Chem. Center, Md.
Edward P. Hardy, Jr., AEC, HASL, N. Y., N. Y.
John H. Harley, AEC, HASL, N.Y., N. Y.
Howard A. Hawthorne, Univ. of Calif., Los Angeles, Calif.
Thomas J. Haycock, AEC, Wash., D. C.
Don W. Hayne, U. S. Fish & Wildlife Service, Laurel, Md.
Robert P. Hayward, USPHS, Wash., D. C.
Walter R. Heald, USDA, Beltsville, Md.
Edward E. Held., Univ. Washington, Seattle, Wash.
Paul S. Henshaw, AEC, Wash., D. C.
Karl E. Herde, AEC, Savannah River Oper. Off., Aiken, S. C.
Carlton M. Herman, U. S. Fish & Wildlife Service, Laurel, Md.
George M. Hidy, Johns Hopkins Univ., Baltimore, Md.
Albert W. Hilberg, USPHS, Wash., D. C.
Lt. Col. John A. Hilchen, Off. Surgeon Gen., U. S. Army R & D Cmd., Wash., D. C.
Joshua Z. Holland, AEC, Wash., D. C.
Hal Hollister, USPHS, Wash., D. C.
John R. Horan, AEC, Idaho Oper. Off., Idaho Falls, Ida.
Charles L. Hosler, Penna. State Univ., University Park, Penna.
Andrew P. Hull, BNL, Upton, L. I., N. Y.
Paul James, USDA, Beltsville, Md.
M. L. Janower, USPHS, Wash., D. C.
Herbert Jehle, George Washington Univ., Wash., D. C.
James E. Johnson, E. I. DuPont & Co., Savannah R. Plant, Aiken, S. C.
Sam P. Jones, General Mills Inc., Minneapolis, Minn.
Harry S. Jordan, LASL, Los Alamos, N. Mex.
Robert L. Junkins, Hanford Labs., Richland, Wash.
Marvin I. Kalkstein, Air Force Cambridge Research Labs., Bedford, Mass.

John H. Kane, AEC, Wash., D. C.
Francis Kawahara, U. S. Naval Radiological Defense Lab., San Francisco, Calif.
Alfred W. Klement, Jr., AEC, Wash., D. C.
Harold A. Knapp, AEC, Wash., D. C.
Joseph B. Knox, Univ. of Calif., LRL, Livermore, Calif.
Robert C. Koch, Nuclear Science & Engrg. Corp., Pittsburgh, Penna.
A. B. Kochanski, U. S. Weather Bureau, Wash., D. C.
Paul Kruger, Hazalton-Nuclear Science Corp., Palo Alto, Calif.
Louis A. Krumholz, Univ. of Louisville, Louisville, Ky.
J. Laurence Kulp, Lamont Geol. Observ., Columbia Univ., N. Y., N. Y.
Paul K. Kuroda, Unvi. of Arkansas, Fayetteville, Ark.
Roy E. Kyner, AEC, Wash., D. C.
Donald L. Lambdin, USPHS, Wash., D. C.
Wright H. Langham, LASL, Los Alamos, N. Mex.
Paul A. LaRock, USPHS, Wash., D. C.
Kermit H. Larson, Univ. of Calif., Los Angeles, Calif.
Paul J. Lavik, Western Reserve Univ., Cleveland, O.
Duane E. Lea, U. S. Navy Weather Research Lab., Norfolk, Va.
Leon Leventhal, Fracerlab Inc., Richmond, Calif.
Robert J. List, U. S. Weather Bureau, Wash., D. C.
Capt. E. A. Lloyd, Nuclear Defense Lab., Army Chem. Center, Md.
R. K. Longaker, AEC, Wash., D. C.
William E. Lotz, AEC, Wash., D. C.
S. Allan Lough, AEC, Wash., D. C.
John F. Loutit, Med. Research Council Radiobiological Research Unit, Harwell, Didcot, Berks., England
R. H. Lowry, The Boeing Co., Seattle, Wash.
William J. McAnally, Jr., USPHS, Wash., D. C.
Tommy F. McCraw, U. S. Air Force (AFCIN-3K2), Wash., D. C.
Lester Machta, U. S. Weather Bureau, Wash., D. C.
James E. Manson, Air Force Cambridge Research Labs., Bedford, Mass.
Richard S. Mark, USPHS, Wash., D. C.
Michael Markels, Jr., Atlantic Research Corp., Alexandria, Va.
William F. Marlow, AEC, Wash., D. C.
Edward A. Martell, Air Force Cambridge Research Labs., Bedford, Mass.
W. L. Marter, E. I. duPont & Co., Savannah R. Plant, Aiken, S. C.
Russell A. Martin, U. S. Air Force (AFCIN-3K2), Wash., D. C.
Ralph B. Mason, U. S. Weather Bureau, Wash., D. C.
Everett R. Mathews, AEC, Albuquerque Oper. Off., Albuquerque, N. Mex.
Homer E. Menker, AEC, HASL, N. Y. N. Y.
Samuel Merrill, USPHS, Wash., D. C.
Milton W. Meyer, USDA, Beltsville, Md.
Irving Michelson, Consumers Union, Mt. Vernon, N. Y.
Glenn H. Miller, Sandia Corp., Albuquerque, N. Mex.
James W. Miller, USPHS, Wash., D. C.
Capt. R. P. Minx, Defense Atomic Support Agency, Wash., D. C.
Norman Bodine, USPHS, Wash., D. C.
Kark Z. Morgan, ORNL, Oak Ridge, Tenn.

Alvin L. Morris, U. S. Navy Weather Research Facility, Norfolk, Va.
James L. Mortensen, Ohio State Univ., Columbus, O.
Donald L. Myhre, USDA, Beltsville, Md.
Norman Myrick, Milk Industry Foundation, Wash., D. C.
Lt. Col. Russell J. Nash, Military Liaison Comm., DOD/AEC, Wash., D. C.
John V. Nehemias, AEC, Wash., D. C.
Reginald E. Newell, Mass. Inst. of Tech., Cambridge, Mass.
Julian M. Nielson, Hanford Labs., Richland, Wash.
H. Nishita, Univ. of Calif., Los Angeles, Calif.
Lt. Col. Max M. Nold, Defense Atomic Support Agency, Wash., D. C.
Thomas F. O'Leary, AEC, Wash., D. C.
John R. Olive, American Inst. Biological Sciences, Wash., D. C.
Jerry S. Olson, ORNL, Oak Ridge, Tenn.
William S. Osburn, Univ. of Colorado, Boulder, Colo.
Roy Overstreet, Univ. of Calif., Berkeley, Calif.
Donald H. Pack, U. S. Weather Bureau, Wash., D. C.
D. I. Packham, British Embassy, Wash., D. C.
Robert L. Patterson, Jr., U. S. Naval Research Lab., Wash., D. C.
Norman E. Polster, Southhampton, Penna.
Robert Porter, USPHS, Wash., D. C.
Capt. T. D. Potter, U. S. Air Force Climatic Center, Wash., D. C.
Capt. William G. Powell, Nuclear Defense Labs. Army Chem. Center, Md.
Samuel C. Rainey, U. S. Naval Radiological Defense Lab., San Francisco, Calif.
Jack W. Reed, Sandia Corp., Albuquerque, N. Mex.
William H. Regan, AEC, Wash., D. C.
William L. Reichel, U. S. Fish & Wildlife Service, Laurel, Md.
Robert F. Reitemeier, AEC, Wash., D. C.
Theodore R. Rice, Bureau Commercial Fisheries, Beaufort, N. C.
Joseph Rivera, AEC, HASL, N. Y. N. Y.
Howard Roberts, Jr., USDA, Beltsville, Md.
James S. Robertson, Bnl, Upton, L. I., N. Y.
Evan M. Romney, Univ. of Calif., Los Angeles, Calif.
Howard Rosenbaum, USPH, Wash., D. C.
Jan Rosinski, Armour Research Foundation, Chicago, Ill.
Col. Irving J. Russell, Air Force Special Weapons Center, Kirtland AFB, N. Mex.
John H. Rust, Univ. of Chicago, Chicago, Ill.
Leonard P. Salter, AEC, HASL, N. Y., N. Y.
Irving H. Schlafman, USPHS, Wash., D. C.
Vincent Schultz, AEC, Wash., D. C.
Robert K. Schulz, Univ. of Calif., Berkeley, Calif.
Jacob Sedlet, ANL, Argonne, Ill.
Lloyd R. Setter, R. A. Taft Sanitary Engrg. Center, USPHS, Cincinnati, O.
Lora M. Shields, New Mexico Highlands Univ., Las Vegas, N. Mex.
James N. Shiveley, USPHS, Wash., D. C.
Lt. Richard L. Showen, Nuclear Defense Labs., Army Chem. Center, Md.
Robert H. Shumway, USPHS, Wash., D. C.
Leslie Silverman, Harvard Univ., Boston, Mass.

Kenneth A. Simmons, AEC, Wash., D. C.
Irving A. Singer, BNL, Upton, L. I., N. Y.
Walter S. Snyder, ORNL, Oak Ridge, Tenn.
Samuel Sperling, USPHS, Wash., D. C.
William C. Spreen, U. S. Air Force Climatic Center, Wash., D. C.
Dean O. Staley, Univ. of Arizona, Tucson, Ariz.
Maj. Albert K. Stebbins, III, Defense Atomic Support Agency, Wash., D.C.
Harold Stephens, Health Physics Off., Army Chem. Center, Md.
Sidney C. Stern, General Mills, Inc., Framingham, Mass.
Lorin R. Stieff, Hq. U. S. Air Force, Wash., D. C.
Conrad P. Straub, R. A. Taft Sanitary Engrg. Cen., USPHS, Cincinnati, O.
Melvin P. Stulberg, AEC, Wash., D. C.
Kosta Telegadas, U. S. Weather Bureau, Wash., D. C.
Sidney Teweles, U. S. Weather Bureau, Wash., D. C.
L. L. Thatcher, U. S. Geological Survey, Wash., D. C.
John C. Thompson, Jr., Cornell Univ., Ithaca, N. Y.
Paul C. Tompkins, USPHS, Wash., D. C.
Robert C. Tompkins, Nuclear Defense Labs., Army Chem. Center, Md.
Lawrence W. Tuttle, Univ. of Rochester, Rochester, N. Y.
Lester Van Middlesworth, Univ. of Tenn., Memphis, Tenn.
Maj. Arthur F. Vetter, Hq. U. S. Air Force, Wash., D. C.
John C. Villforth, USPHS, Wash., D. C.
Herbert L. Volchok, Isotopes,Inc., Westwood, N. J.
Ray D. Walton, Jr., AEC, Wash., D. C.
B. H. Webb, USDA, Wash., D. C.
(Mrs.) Audrey Wegst, Univ. of Michigan, Ann Arbor, Mich.
E. S. Weiss, USPHS, Wash., D. C.
Ira B. Whitney, AEC, HASL, N. Y., N. Y.
Dale E. Wilkins, USDA, Beltsville, Md.
Lt. Col. George M. Williams, Hq. U. S. Air Force, Wash., D. C.
Edward F. Wilsey, Nuclear Defense Labs., Army Chem. Center, Md.
John N. Wolfe, AEC, Wash., D. C.
Rex C. Wood, General Mills Inc., Minneapolis, Minn.
Lt. Col. Gilbert N. Woods, Hq. U. S. Air Force, Wash., D. C.

AEC - U. S. Atomic Energy Commission
ANL - Argonne National Laboratory
BNL - Brookhaven National Laboratory
HASL - Health and Safety Laboratory
LASL - Los Alamos Scientific Laboratory
ORINS - Oak Ridge Institute of Nuclear Studies
ORNL - Oak Ridge National Laboratory
USDA - U. S. Dept. of Agriculture
USPHS - U. S. Public Health Service
SFOO - San Francisco Operations Office