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FALLOUT PROGRAM
QUARTERLY SUMMARY REPORT

January 1, 1972

UNITED STATES ATOMIC ENERGY COMMISSION
NEW YORK, N. Y. 10014

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HEALTH AND SAFETY LABORATORY

FALLOUT PROGRAM
QUARTERLY SUMMARY REPORT

(September 1, 1971 through December 1, 1971)

Prepared by

Edward P. Hardy, Jr.

Environmental Studies Division

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January 1, 1972

UNITED STATES ATOMIC ENERGY COMMISSION
Health and Safety Laboratory
New York, N. Y.

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FALLOUT PROGRAM
QUARTERLY SUMMARY REPORT

January 1, 1972

This report presents current data from the HASL Fallout Program, the Radiological Physics and Chemistry Divisions at Argonne National Laboratory, the Air Resources Laboratories of the National Oceanic and Atmospheric Administration, and the EURATOM Joint Nuclear Research Centre at Ispra, Italy. The initial section consists of interpretive reports and notes covering the following topics: airborne Pu-239 at Rocky Flats, C-14 in the stratosphere, a high-volume air ejector sampler, stable lead in New York City diet, Sr-90 in diet, and quality control analyses at HASL. Subsequent sections include tabulations of radionuclide levels in fallout, surface air, milk, and tap water. A bibliography of recent publications related to radionuclide studies, is also presented.

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INTRODUCTION

Every three months, the Health and Safety Laboratory issues a report summarizing current information obtained at HASL pertaining to fallout. This report, the latest in the series, contains information that became available during the period from September 1, 1971 to December 1, 1971. The next report is scheduled for publication April 1, 1972. Preceding reports in the series, starting with HASL-42, "Environmental Contamination from Weapons Tests", and continuing through HASL-246, (this report) may be purchased from the Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Virginia 22151. A complete listing of these Fallout Program Quarterly Summary Reports is given on the title page of this report.

To give a more complete picture of the current fallout situation and to provide a medium for rapid publication of radionuclide and trace element data, these quarterly reports often contain information from other laboratories and programs, some of which are not part of the general AEC program. To assist in developing, as rapidly as possible, provisional interpretations of the data, special interpretive reports and notes prepared by scientists working in the field of fallout are also included from time to time. Many of these scientists are associated in some way with the general AEC program. Information developed outside of HASL is identified as such and is gratefully acknowledged by the Laboratory. In this report, data from the EURATOM Joint Nuclear Research Centre at Ispra, Italy, the Divisions of Radiological Physics and Chemistry at Argonne National Laboratory, and the Air Resources Laboratories of the National Oceanic and Atmospheric Administration, are given.

A portion of the radiochemical analyses either have been or are being carried out by commercial laboratories under contract to the HASL Environmental Studies Division. The results of these analyses are reported as part of HASL's regular fallout program. The contractor analytical laboratories which provided data are Nuclear Science and Engineering Corporation, Pittsburgh, Pa.; Isotopes, Inc., Westwood, N.J.; Radiochemistry Incorporated, Louisville, Ky; Tracerlab, Div. of LFE, Richmond, Calif. (now Trapelo Div./West); Controls for Radiation, Inc., Cambridge, Mass; Hazleton-Nuclear Science Corp., Palo Alto, Calif. (now Isotopes Palo Alto Laboratories); Food, Chemical and Research Laboratories, Inc., Seattle, Washington; Custom Nuclear Co., Mountainview, Calif., Ledoux and Company, Teaneck, N. J., and U. S. Testing Co., Richland, Washington.

This report is divided into four main parts:

1. Interpretive Reports and Notes
2. HASL Fallout Program Data
3. Data from Sources Other than HASL
4. Recent Publications Related to Radionuclide Studies

PART I
INTERPRETIVE REPORTS
and
NOTES

X PLUTONIUM IN THE NEIGHBORHOOD OF ROCKY FLATS, COLORADO:
AIRBORNE RESPIRABLE PARTICLES

by H. L. Volchok, (HASL)
R. Knuth, (HASL)
M. T. Kleinman, (HASL)

ABSTRACT

Several experiments were carried out, off-site, but close to the Rocky Flats plutonium plant, to determine the respirable fraction of the resuspended Pu^{239} . The results indicate that in this area, with concentrations in excess of 1 fCi/m^3 , the respirable fraction is about 0.2 to 0.4. These experiments also provided evidence that the main source of the resuspended Pu^{239} is probably located somewhat east of the plant site, and that very steep vertical gradients of Pu concentrations may be present in the air in this vicinity.

Introduction and Background

The Rocky Flats plant, operated by the Dow Chemical Company for the Atomic Energy Commission, produces plutonium parts for nuclear weapons. It is located about 20 miles northwest of Denver and 12 miles south of Boulder, Colorado. A number of other towns of 2,000 or more population are within 8 to 10 miles to the north, east and south.

National interest in the environmental situation with regard to plutonium, in the area near the Rocky Flats plant, was aroused in early 1970. Dr. Edward Martell, at that time representing the Colorado Committee for Environmental Information (CCEI), demonstrated the presence of excess plutonium in soil near the plant (1). The CCEI investigation, spurred by a serious fire at the plant in May of 1969, suggested among other things, that curies to tens of curies of plutonium from the plant had been deposited in offsite areas, and that "...the continuation of plant operations at this site represents a serious threat to the health and safety of the people of the Denver area."

The Health and Safety laboratory (HASL) conducted a study of the distribution of Pu^{239} in the offsite neighborhood of the plant, in February 1970. This study has been completed and published, (2). The major findings were:

- (a) The most likely source of the offsite plutonium contamination was barrels of contaminated cutting oil, stored on the southeast corner of the plant property, and known to have leaked.

- (b) The upper limit of the inventory of Pu^{239} on non government property, which could be attributed to contamination from the Rocky Flats plant was 5.8 curies.
- (c) The pattern of contamination on the ground is generally compatible with the average wind vectors for the area. This, along with the low precipitation and high winds, suggests that the major mechanism of transport of the plutonium from the drum storage area, was resuspension.

In line with this last conclusion, and reflecting the understanding that inhalation (in contrast to ingestion) is probably the more effective mode of entry of plutonium into critical organs (3,4), HASL, in mid 1970, set up air sampling equipment at the east boundary fence of the plant. This point is adjacent to the barrel storage area. Our motive was to systematically document the Pu^{239} concentration in the air in the area of highest ground concentration. The results (5) indicate that the Pu^{239} levels, while quite variable, generally correlated with wind velocities of over 8 miles per hour. The monthly concentrations

averaged over ten times those expected from fallout, and varied between 1 and 10% of the most conservative maximum permissible concentration (mpc), which is 60 fCi/m³ for soluble Pu with bone as the critical organ.

This report describes our most recent experiments at Rocky Flats, attempting to characterize the size of the plutonium bearing particles in the aerosol. This information bears directly on the facility for these particles to penetrate the respiratory tract and lungs, and therefore may be used to evaluate the radiation risk. The usual ways of estimating the hazard due to insoluble particles depend upon a knowledge of that portion of the inhaled dust which is deposited in the non-ciliated part of the lung. This is termed the "respirable" fraction (6,7,8).

The British Medical Research Council (BMRC) defined the characteristics of the respirable fraction of inhaled dust in terms of the aerodynamic diameter (AD) of the particles (7). Hence, all particles of diameter less than 1.5 microns (μm) are respirable, while only 50% of the 5.0 μm and none larger than 7.1 μm are deposited.

At a conference at the Los Alamos Scientific Laboratory (LASL) in 1961, a slightly different set of particle sizes were

developed, to define the respirable fraction (6). In this, the deposited percentages are: 100% below $2\mu\text{m}$, 50% of $3.5\mu\text{m}$ and 0% over $10\mu\text{m}$.

A simple mathematical relationship between the two lung retention models has been developed (9,10,11) as follows:

$$\text{FR}_{\text{LASL}} = 0.8 (\text{FR}_{\text{BMRC}}) \pm 0.1 (\text{FR}_{\text{BMRC}})$$

In this FR_{LASL} = the fraction respirable by the LASL model,

FR_{BMRC} = the fraction respirable by the BMRC model.

Hence, the BMRC assumptions will result in slightly higher respirable fractions than the LASL.

The only information directly pertaining to this specific problem at Rocky Flats was developed in a study of particles on the HASL filter papers (12). In this, the Pu^{239} was found on "host" particles with a mean diameter of about $10\mu\text{m}$. From the particle size distribution presented, and assuming a density of 2.5 for the particles, the respirable fractions were calculated to be between .05 and .07.

Sampling and Analysis

The sampling was carried out during the period October 12 - 14, 1971 near Rocky Flats. Our primary goal was to assess the respirable fraction of the airborne Pu^{239} in terms of both the BMRC and LASL models. The BMRC definition can be approximated by use of a horizontal elutriator followed by a high efficiency filter (13,14,15). The elutriator used in these tests is 20 cm. wide, 50 cm. long with 19 channels spaced at 0.124 cm. It was operated at a flow rate of 6.25 cubic feet per minute ($= 0.175 \text{ m}^3$ per minute) to give 100% removal efficiency for $7.1 \mu\text{m}$ particles as specified in the BMRC model. The respirable fraction passing through the elutriator was filtered on Microsorban, a highly efficient polystyrene material.

The LASL model can also be approximated by a two stage sampler consisting of a cyclone and filter (6,16). We used an instrument essentially described in reference (6); a one inch diameter cyclone first stage operated at a flow rate of 3.5 cubic feet per minute ($= 0.098 \text{ m}^3$ per minute) backed again by a Microsorban filter for the respirable fraction.

The immediate neighborhood of Rocky Flats with the locations of the four sampling sites, is shown in Fig. 1. The east-southeast

Fig. 1 - The Environs of Rocky Flats Plant, Colorado

sampling pattern was chosen because of the prevailing wind patterns and the observed distribution of Pu^{239} in the soil (2). The "S" numbers designating the stations are those used by Dow in their environmental surveillance reports (e.g., 17). S-8 is just inside of the east security fence, very close to the point of continuous air sampling mentioned above (5). This site is considered most important and was therefore most heavily instrumented. The elutriator (only one was available) and two cyclone sets were all run continuously for about 54 hours at S-8. In addition, two hi-vol Staplex samplers, with Microsorban filters were run for the same period as a control, to measure the total Pu^{239} air concentration. All of the samplers at this site were operated at about 3 feet above the ground.

At the other three sites conditions were much less ideal due to power shortage and a number of compromises had to be made. At S-32 and S-34 we could only run one cyclone for the entire period and one Staplex for about 70% of the time. Further, the cyclones were located at about 3 ft. above the ground while the Staplex was at about 7.5 ft. At site 18, the electrical power was even more restricted, and only one cyclone could be operated.

The winds were quite variable through most of the sampling period. During the first day they were mainly from the north and northwest, and averaged about 6 miles per hour (mph), with gusts up to 33 mph. The second day had mostly southerly and "variable" winds reported, again averaging 6 and gusting to 21 mph. On the third day we experienced the prevailing wind patterns, mostly NW, W and SW. These winds averaged 11 mph and there was a westerly gust of 43 mph.

The plutonium analyses were carried out by both HASL and Trapelo/West. Both laboratories received "blind" knowns and blanks for evaluation of the quality of the analyses. In all cases the blanks were less than 0.01 dpm Pu^{239} and the knowns were reported to closer than 10% of the true value.

Results and Discussion

Table 1 lists the results from the four sampling sites near the Rocky Flats plant. The samples from site-8, near the east gate of the plant, are very consistent with respect to both total Pu^{239} concentration and the respirable fraction. The mean concentration for the total 54 hour sampling period, based upon 4 data (two staplex and two cyclone sets) was $6.25 \pm 0.35 \text{ fCi/m}^3$. The average

Table 1

Pu²³⁹ CONCENTRATIONS IN ROCKY FLATS PARTICLE SAMPLING EXPERIMENT

<u>Site</u>	<u>Sample</u>	<u>Volume (m³)</u>	<u>Pu²³⁹ (fCi/m³) *</u>	<u>Fraction Respirable</u>
S-8	Elutriator Filter	566	1.82	0.29**

	Cyclone-6	318	4.55	
	Cyclone-6 Filter	318	0.80	0.15

	Cyclone-4	316	5.19	
	Cyclone-4 Filter	316	1.42	0.21
S-32	-----			
	Staplex-A	4097	6.64	
	Staplex-B	4226	6.38	

	Cyclone-1	316	12.34	
	Cyclone-1 Filter	316	6.58	0.35

S-18	Staplex-C	2957	5.80	

	Cyclone-2	295	0.10 (A)	
S-34	Cyclone-2 Filter	295	0.07 (A)	-

S-34	Cyclone-3	313	1.38	
	Cyclone-3 Filter	313	0.12 (A)	0.08 (A)

	Staplex-D	4256	0.03 (A)	

* 1 σ errors are less than $\pm 10\%$ unless noted by.

(A) for errors of $\pm 50\%$ or greater.

** calculated by use of average total concentration = 6.25 ± 0.35 fCi/m³.

respirable fraction from the two cyclone experiments was 0.18. Using the relationship described earlier, between the BMRC and LASL models, and based on the cyclone result, we would expect the elutriator experiment to have indicated a respirable fraction of $0.23 \pm .03$. In fact the elutriator filter, when compared with the average concentration during the period indicates 0.29 as the respirable fraction.

At site S-32 the results were quite surprising in that the combined cyclone and filter indicate a Pu^{239} concentration about three times that found at the closer in S-8 site. The Staplex, at S-32 did not come out in agreement with the cyclone set, but this may be due to the abbreviated sampling period, and the difference in sampler height, as described earlier. The respirable fraction was 0.35.

Although the results from sites S-18 and S-34 were too low to give meaningful respirable fractions, the activity found in the S-34 cyclone suggests that the Pu^{239} concentration in the air there was about an order of magnitude higher than at S-18. The Staplex again, as at S-32 was much lower in concentration.

Not too many conclusions can be drawn from so few data. The respirable fraction of Pu^{239} in the air near the Rocky Flats plant seems to be reasonably consistent at about 0.2 to 0.4. This is not in very close agreement with the value of .05 to .07 derived from particle size measurements on filter papers, described earlier (12). Although there is no direct proof at this time, we feel that this discrepancy is due mainly to a discrimination against smaller particles, in the filter paper work. In the filters studied very few particles of less than $2\text{ }\mu\text{m}$ were found to contain plutonium. Whether this is due to the actual method of particle sizing, cannot be determined. In any case, this sort of bias would lead to lower respirable fractions, as observed.

The 0.2 to 0.4 respirable fraction determined in this study, is not too different from the value assumed in the derivation of the maximum permissible concentration (18). Hence the concentrations observed can be compared with the published mpcs. The highest value during the period of this experiment, almost 19 fCi/m^3 at S-32, was more than 30% of the most conservative guideline for general population exposure mentioned earlier.

The mean concentrations seen at the two highest level sites suggest that the major source of resuspended plutonium is probably not within the plant site, but somewhere between S-8 and S-32, although the wind patterns during these tests makes this conclusion somewhat tenuous. Finally, in comparing the Staplex results with the cyclone + filter at the two stations S-32 and S-34 we believe that there must be a substantial vertical gradient in Pu^{239} concentration, at least in the range of 3 to 8 feet above the ground. The agreement at S-8, where the samplers were all at about the same elevation reinforces this very tentative conclusion.

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ANALYSES OF QUALITY CONTROL SAMPLES AT HASL AND A
CONTRACTOR LABORATORY DURING 1971

By E. Hardy (HASL)

ABSTRACT

Samples of biological material and fallout, analyzed at HASL and by a contractor laboratory include quality control samples which are submitted as blinds. The checks consist of blanks, natural samples analyzed repeatedly from year to year, replicates or splits of unknown, spikes, and duplicate samplings. This report summarizes quality control data for ashed bone, diet and fallout samples analyzed for Sr^{90} , Sr^{89} , Cs^{137} , and Ca during 1971.

INTRODUCTION

Analyses performed either at HASL or by contractor laboratories for HASL, include samples that are used to assess the quality of the data. Each group of unknowns submitted for analysis consists of check samples which constitute about ten percent of the total number of samples.

Quality control results for stratospheric and surface air samples are regularly presented in other sections of this Quarterly report. Results for check samples submitted with monthly fallout collections, diet and human bone samples,

however, are presented on an annual basis (1,2). This report covers these types of quality control samples analyzed during 1971 as well as results of duplicate samplings of tap water and monthly fallout.

The actual samples used in quality control consist of blanks, natural samples analyzed repeatedly over several years, replicates or splits of unknowns, and spiked samples. In the accompanying tables, samples analyzed at the Health and Safety Laboratory are identified as HASL while those analyzed at a contractor laboratory are identified by the code letters (NN).

Following is a summary of quality control data for bone ash, vegetation ash, ion exchange resin and simulated pot samples analyzed during 1971. Results of individual analyses and average values for 1. blanks, 2. natural samples, 3. duplicates and 4. spikes, are given in tables 1 through 4. Results for duplicate sample collections are presented in table 5.

SUMMARY OF RESULTS

1. Blanks

Fourteen blind calcium phosphate samples (10 grams each) were analyzed for Sr^{90} at HASL during 1971. The average value was 0.05 ± 0.05 dpm Sr^{90} per gram. The error term is one standard

deviation from the mean. This average value is the same as that reported in 1970.

Seventy gram aliquots of Dowex 50W-X12, 50-100 mesh, cation exchange resin, conditioned at HASL and used in monthly fallout collections employing a funnel and ion-exchange column system, were analyzed as blanks. Contractor laboratory NN analyzed 17 unexposed resin samples during the course of the FY'71 contract period and reported, on the average, 0.8 ± 1.3 dpm Sr^{90} . Twelve resin blanks were analyzed at HASL and the average during 1971 was 0.5 ± 0.3 dpm Sr^{90} . These values are not significantly different from those reported in 1970. These blank levels are considered acceptable in terms of the activity level (~ 2 dpm) for a column that has received a deposit of 0.01 mCi Sr^{90} per km^2 which is the lowest monthly rate reported. The simulated pot collections analyzed as blanks showed similar Sr^{90} levels. The average of 17 analysis by contractor NN was 0.6 ± 0.9 dpm Sr^{90} while the average of 4 analyses at HASL was 0.6 ± 0.6 dpm Sr^{90} .

Blank resin and simulated pot samples analyzed for Sr^{89} in 1971 by contractor laboratory NN averaged 0.9 ± 2.2 and 0.4 ± 0.3 dpm, respectively. The mean and standard deviation for the blank resin are higher than reported in 1970 due to one anomalously high value.

The 1971 blank data are summarized in the following table:

<u>Matrix</u>	<u>Lab.</u>	<u>No. of Analyses</u>	<u>BLANKS</u>		<u>Avg. dpm Sr⁸⁹ per sample</u>
			<u>Avg. dpm Sr⁹⁰ per g</u>	<u>per sample</u>	
Calcium phosphate	HASL	14	0.05±0.05		
Resin	NN	17		0.8±1.3	
	HASL	12		0.5±0.3	
	NN	15			0.9±2.2
Pot	NN	17		0.6±0.9	
	HASL	4		0.6±0.6	
	NN	10			0.4±0.3

2. Natural Samples

Ten grams aliquots of ashed natural samples of human and animal bone, milk, and vegetation have been analyzed for Sr⁹⁰ and Ca repetitively at HASL for a number of years. The following table compares the average of values reported in 1971 with the average of all results for each sample to the end of 1971. The error term accompanying each result is one standard deviation of the mean. Most of these samples have been used in quality control at HASL for ten or more years.

NATURAL SAMPLES (HASL)

<u>Matrix</u>	Orig. <u>HASL No.</u>	<u>Avg. dpm Sr⁹⁰ per g*</u>	
		<u>1970</u>	<u>All analyses</u>
Human bone ash	5407	1.4±0.2 (6)	1.3±0.1 (97)
Milk ash	7604	2.3±0.1 (2)	2.2±0.2 (77)
	9598	1.4 (1)	2.0±0.3 (9)
Vegetation ash	9236	8.2 (1)	9.2±0.8 (21)
	V0169	12.7±0.1 (3)	13.3±1.0 (32)
	V0360	16.6±0.4 (3)	16.0±1.1 (13)

*No. of analyses in parenthesis

In two cases (milk ash sample No. 9598 and vegetation ash sample No. 9236) the 1971 average is lower than the average of all available analyses, considering the limits of error. For the other samples the two values agree within the limits of error reported. All values have been corrected for decay to a common date assigned to each sample.

3. Duplicates

Split blind duplicate human bone and food ash samples were analyzed for Sr⁹⁰ at HASL during 1971. Average percent standard deviations from the means were less than ten percent as shown in the following table:

BLIND DUPLICATES (HASL)

<u>Matrix</u>	No. of sets of dupl. <u>analyses</u>	Avg. % std. dev. from mean	
		<u>Sr⁹⁰</u>	<u>Ca</u>
Human bone ash	11	9.7	
	10		1.4
Food ash	16	7.0	

The overall analytical reproducibility is considered satisfactory for these types of samples and activity levels.

A. Spikes

Known amounts of standardized Sr⁹⁰ and Sr⁸⁹ solutions are added to simulated pot and unexposed resin samples and submitted with unknowns to HASL and the contractor laboratory. Average recoveries, activity found ÷ activity added ×100, for analyses performed during 1971 are summarized below. The error term is one Gaussian standard deviation from the mean.

SPIKED SAMPLES

<u>Type sample</u>	No. of <u>pairs</u>	<u>% Sr⁹⁰ recovered</u>	
		<u>Lab. NN</u>	<u>HASL</u>
Resin	35	79±19	
	12		102±7
Pot	34	80±17	
	7		97±11

<u>Type sample</u>	<u>No. of pairs</u>	<u>% Sr⁸⁹ recovered (Lab. NN) (activity level-dpm)</u>		
		<u>10-20</u>	<u>100-200</u>	<u>500-1000</u>
Resin	6 each	84±30	90±27	92±14
Pot	5	90±40		
	6 each		92±27	72±24

Average recoveries of Sr⁹⁰ by the contractor laboratory during 1971 continued to be low as in 1970. A study of this problem is currently underway.

Recovery values for Sr⁸⁹ in the 10-20 dpm range are less variable than in 1970. The relatively larger error terms associated with the mean recovery values for Sr⁸⁹ indicate the inherently less precise procedure of determining Sr⁸⁹ by difference. In general the Sr⁸⁹ recoveries over the 10-1000 dpm range are considered acceptable for the program.

5. Duplicate Sampling

For many years duplicate fallout collectors have been exposed for monthly periods on the roof of HASL to test sampling reproducibility. Duplicate tap water samples from the HASL have also been collected daily and composited on a monthly basis, for the same purpose. Analytical values for duplicate sample collections in 1971 which are given in Tables 5A and 5B may be summarized as follows:

DUPLICATE MONTHLY SAMPLE COLLECTIONS

<u>Sample type</u>	<u>No. of pairs</u>	<u>Avg. % dev. fr. mean</u>		<u>Range</u>	
		<u>Sr⁹⁰</u>	<u>Cs¹³⁷</u>	<u>Sr⁹⁰</u>	<u>Cs¹³⁷</u>
Fallout	9	2.8		0-13%	
	10		4.1		0.5-5.5%
Tap water	11	3.4		0-11%	

In both cases sampling reproducibility is less than five percent.

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HASL-217, p. I-130, January (1970)
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1. BLANKS

A. Calcium Phosphate, Sr⁹⁰

Data Period: January - December 1971

Laboratory: HASL

dpm Sr⁹⁰
per gram

≤0.03

0.03

≤0.04

≤0.01

0.06

≤0.03

≤0.02

≤0.02

0.19

0.06

0.02

0.09

0.09

0.06

14 analyses: avg. 0.05 ± 0.05 dpm Sr⁹⁰ per gram

*A less than or equal to sign (≤) precedes the Poisson standard deviation in those cases where the value obtained was less than the error term or less than zero. For purposes of averaging, the less than or equal sign was disregarded.

1. BLANKS (cont'd)

B. Ion Exchange Resin, Sr⁸⁹ and Sr⁹⁰

Data Period: December 1970 - November 1971: Lab NN
January 1971 - December 1971: HASL

---- dpm Sr ⁹⁰ --		dpm Sr ⁸⁹
<u>Lab NN</u>	<u>HASL</u>	<u>Lab NN</u>
1.2	≤ 0.2	≤ 0.2
1.8	0.6	≤ 0.1
2.0	≤ 0.2	≤ 0.3
≤ 0.3	≤ 0.2	≤ 0.1
≤ 0.1	≤ 0.2	≤ 0.3
5.4	≤ 0.2	≤ 0.1
≤ 0.2	1.1	≤ 0.3
≤ 0.1	0.6	≤ 0.9
≤ 0.1	1.0	≤ 0.3
≤ 0.3	0.6	≤ 0.5
0.3	0.5	8.9
≤ 0.3	<u>0.8</u>	≤ 0.1
0.3	12 analyses	≤ 0.4
≤ 0.2	avg. 0.5±0.3 dpm Sr ⁹⁰	≤ 0.3
≤ 0.1		≤ 0.1
≤ 0.3		
<u>≤ 0.3</u>		
17 analyses		15 analyses
avg. 0.8±1.3 dpm Sr ⁹⁰		avg. 0.9±2.2 dpm Sr ⁸⁹

1. BLANKS (cont'd)

C. Simulated Pot Collection, Sr⁸⁹ and Sr⁹⁰

Data Period: December 1970 - November 1971: Lab NN
 January 1971 - December 1971: HASL

----- dpm Sr ⁹⁰ ----		dpm Sr ⁸⁹
<u>Lab NN</u>	<u>HASL</u>	<u>Lab NN</u>
≤0.3	≤0.2	≤0.1
≤0.2	0.7	≤0.1
≤0.3	1.4	≤0.3
≤0.1	≤0.2	≤0.3
≤0.1		≤0.4
1.9	4 analyses	≤0.3
≤0.1	avg. 0.6±0.6 dpm Sr ⁹⁰	≤0.1
≤0.2		≤0.7
≤0.1		≤0.9
1.4		≤0.5
≤0.1		
2.1		10 analyses
3.1		avg. 0.4 ± 0.3 dpm Sr ⁸⁹
≤0.3		
≤0.3		
≤0.1		
≤0.2		
17 analysis		
avg. 0.6±0.9 dpm Sr ⁹⁰		

2. NATURAL SAMPLES

A. Human Bone Ash (Orig. HASL No. 5407), Sr^{90} and Ca

Data Period: January - December 1971

Laboratory: HASL

<u>dpm Sr^{90}</u> <u>per gram(1)</u>	<u>% Ca</u>
1.24	37.5
1.31	37.0
1.51	37.4
1.76	37.4
1.17	36.2
1.33	37.0

(1) decay corrected to 7/1/57

Avg. of results from July 1957 to December 1971 (2)

dpm Sr^{90} per gram: 1.28 ± 0.14 (10.9%) - decay corrected
to 7/1/57

0.89 ± 0.10 - decay corrected
to 1/1/72

based on 97 blind analyses at HASL

% Ca: 36.5 ± 1.1 (3.0%)
based on 43 blind analyses at HASL

(2) error term is one standard deviation
of the mean

2. NATURAL SAMPLES - cont'd)

B. Powdered Milk Ash (Orig. HASL No. 7604), Sr^{90} and Ca

Data Period: January - December 1971

Laboratory: HASL

dpm Sr^{90} (1)

<u>per gram</u>	<u>% Ca</u>
-----------------	-------------

2.19	14.5
------	------

2.32	14.8
------	------

(1) decay corrected to 3/1/59

Avg. of results from March 1959 thru December 1970(2)

dpm Sr^{90} per gram: 2.21 ± 0.22 (10.0%) decay corrected to
3/1/59

1.61 ± 0.16 decay corrected to
1/1/72

based on 77 blind analyses at HASL

% Ca: 15.1 ± 0.6 (4.0%)
based on 47 blind analyses at HASL

(2) error term is one standard deviation of the mean

2. NATURAL SAMPLES - (cont'd)

C. Milk Ash (Orig. HASL No. 9598), Sr^{90} and Ca

Data Period: January - December 1971

Laboratory: HASL

dpm Sr^{90} (1)

per gram

% Ca

1.44

9.5

(1) decay corrected to 5/1/59

Avg. of results from May 1959 thru December 1971 (2)

dpm Sr^{90} per gram: 2.04 ± 0.29 (14.2%) decay corrected
to 5/1/59

1.49 ± 0.21

decay corrected
to 1/1/72

based on 9 blind analyses at HASL

% Ca

9.8 ± 0.4 (4.1%)

based on 4 blind analyses at HASL

(2) error term is one standard deviation of the mean

2. NATURAL SAMPLES (cont'd)

D. Vegetation Ash (Orig. HASL No. 9236), Sr^{90} and Ca

Data Period: January - December 1971

Laboratory: HASL

dpm Sr^{90} (1)
per gram

8.25

(1) decay corrected to 5/1/59

Avg. of results from May 1959 thru December 1971 (2)

dpm Sr^{90} per gram: 9.22 ± 0.82 (8.9%) decay corrected to
5/1/59

6.74 ± 0.60 decay corrected to
1/1/72

based on 21 blind analyses at HASL

% Ca: 9.7 ± 0.3 (3.2%)

based on 12 blind analyses at HASL

(2) error term is one standard deviation of
the mean

2. NATURAL SAMPLES - (cont'd)

E. Vegetation Ash (orig. HASL No. V0169), Sr^{90} and Ca

Data Period: January - December 1971

Laboratory: HASL

dpm Sr^{90} (1)
per gram

12.6

12.9

12.6

(1) decay corrected to 4/1/63

Avg. of results from April 1963 thru December 1971 (2)

dpm Sr^{90} per gram: 13.3 ± 1.0 (7.3%) decay corrected to
4/1/63

10.7 ± 0.8 decay corrected to
1/1/72

based on 32 blind analyses at HASL

% Ca 2.2 ± 0.1 (4.5%)

based on 6 blind analyses at HASL

(2) error term is one standard deviation of the mean

2. NATURAL SAMPLES (cont'd)

F. Vegetation Ash (orig. HASL No. V0360), Sr^{90}

Data Period: January - December 1971

Laboratory: HASL

dpm Sr^{90} (1)
per gram

16.2

16.9

16.8

(1) decay corrected to 5/1/68

Avg. of results from May 1968 to December 1971 (2)

dpm Sr^{90} per gram: 16.0 ± 1.1 (7.0%) decay corrected to
5/1/68

14.6 ± 1.0 decay corrected to
1/1/72

based on 13 blind analyses at HASL

(2) error term is one standard deviation of
the mean

3. BLIND DUPLICATES

A. Human Bone Ash

<u>HASL No.</u>	<u>dpm Sr⁹⁰ per gram</u>	<u>% Ca</u>
B2362	0.67	38.9
B2370	0.58	38.9
B2386	0.40	
B2388	0.29	
B2400	0.42	36.9
B2401	0.44	37.2
B2360	0.56	37.5
B2369	0.58	38.9
B2381	0.58	34.9
B2387	0.56	35.2
HB4379	1.10	36.5
HB4391	1.03	36.4
HB4384	2.20	36.6
HB4392	2.61	36.2
HB4396	0.60	36.3
HB4403	0.70	35.2
HB4400	0.97	37.4
HB4404	0.97	37.8
HB4413	0.57	38.4
HB4430	0.64	38.6
HB4416	0.61	38.4
HB4430	0.64	38.6

11 Sets of Sr⁹⁰ blind duplicates

Avg. % Standard deviation from mean: 9.7% range: 0 - 32%

10 sets of Ca blind duplicates

Avg. % Standard deviation from mean: 1.4% range: 0 - 3.6%

3. BLIND DUPLICATES (Cont'd)

B. Vegetation and Food Ash

Data Period: January - December 1971

Laboratory: HASL

<u>HASL No.</u>	dpm Sr ⁹⁰ <u>per gram</u>	<u>HASL No.</u>	dpm Sr ⁹⁰ <u>per gram</u>
F2789	2.72	F2882	3.54
F2807	2.77	F2900	3.26
F2795	1.23	F2888	0.86
F2808	1.26	F2901	0.90
F2811	0.56	F2904	0.47
F2829	0.70	F2922	0.43
F2817	0.63	F2910	1.25
F2830	0.64	F2923	1.12
F2833	5.45	F2926	4.39
F2851	5.24	F2944	4.39
F2839	1.37	F2932	1.41
F2852	1.20	F2945	1.57
F2855	0.98	F2952	0.61
F2873	1.06	F2970	0.69
F2861	0.42	F2958	1.22
F2874	0.42	F2971	1.26

16 sets of blind duplicates

Avg. % standard deviation from mean: 7.0% range: 0 - 22%

4. SPIKES

A. Ion-Exchange Resin, Sr⁹⁰

Data Period: December 1970 - November 1971: Lab NN
January 1971 - December 1971: HASL

Lab NN dpm Sr ⁹⁰		HASL dpm Sr ⁹⁰	
<u>added</u>	<u>found</u>	<u>added</u>	<u>found</u>
25	14	25	27
25	18	25	26
25	17	25	25
25	23	25	25
25	23	26	29
25	23	26	25
25	9	26	26
25	11	50	49
26	25	50	49
26	26	50	59
26	21	50	50
26	21	50	47
26	24		
26	21		
26	14		
26	24		
26	23		
26	25		
50	45		
50	43		
50	21		
50	40		
50	35		
50	42		
50	50		
50	34		
50	51		
50	26		
50	43		
50	29		
50	31		
50	47		
50	56		
50	45		

12 pairs of results:
avg % recovered: 102±7%

35 pairs of results:
avg. % recovered: 79±19%

4. SPIKES (Cont'd)

B. Simulated Pot Collections, Sr⁹⁰

Data Period: December 1970 - November 1971: Lab NN
 January 1971 - December 1971: HASL

Lab NN dpm Sr ⁹⁰		HASL dpm Sr ⁹⁰	
<u>added</u>	<u>found</u>	<u>added</u>	<u>found</u>
25	23	25	26
25	19	25	25
25	19	26	22
25	16	26	29
25	29	50	41
25	23	50	51
25	23	50	47
26	22	<hr/>	
26	27	7 pairs of results avg. % recovered: 97±11%	
26	15		
26	19		
26	13		
26	23		
26	25		
26	12		
26	24		
50	52		
50	40		
50	36		
50	29		
50	41		
50	26		
50	38		
50	30		
50	48		
50	53		
50	39		
50	38		
50	43		
50	39		
50	40		
50	35		
50	51		
50	45		

34 pairs of results
 avg. % recovered: 80±17%

4. SPIKES (cont'd)

C. Ion-Exchange Resin, Sr⁸⁹

Data Period: November 1970 - November 1971

Lab: NN

dpm Sr ⁸⁹		
<u>added</u>	<u>found</u>	
14	12	
16	10	6 pairs of results
16	9	
16	12	avg. % recovered: 84±30%
17	14	
17	24	

144	161	
151	110	6 pairs of results
159	184	
159	179	avg. % recovered: 90±27%
166	83	
166	131	

755	752	
763	694	6 pairs of results
786	743	
800	829	avg. % recovered: 92±14%
800	518	
808	814	

4. SPIKES (cont'd)

D. Simulated Pot Collections, Sr⁸⁹

Data Period: November 1970 - December 1971

Lab: NN

<u>added</u>	dpm Sr ⁸⁹ <u>found</u>	
14	20	
16	14	5 pairs
16	18	
17	7	avg. % recovered: 90±40%
17	11	

144	190	
151	139	6 pairs of results
159	94	
159	175	avg. % recovered: 92±27%
166	115	
166	147	

755	585	
763	692	6 pairs of results
786	604	
800	685	avg. % recovered: 72±24%
800	601	
808	204	

5. DUPLICATE SAMPLING

A. Tap Water (Monthly Composites)

Data Period: January - December 1971

Laboratory: HASL

<u>HASL No.</u>	<u>dpm Sr⁹⁰</u> <u>per 100 l</u>	<u>dpm Cs¹³⁷</u> <u>per 100 l</u>
W0464	136	19.5
W0465	lost	19.6
W0467	155	22.4
W0468	157	21.1
W0470	153	16.0
W0471	146	16.5
W0472	166	15.1
W0473	166	13.9
W0476	156	12.4
W0477	137	13.1
W0478	lost	14.1
W0479	143	14.6
W0486	142	15.8
W0487	138	15.3
W0492	138	18.9
W0493	140	18.2
W0494	118	
W0495	120	
W0498	113	17.6
W0499	113	18.2
W0500	159	20.9
W0501	160	20.1

9 sets of duplicate samplings for Sr⁹⁰
avg. % deviation from mean: 2.8%, range 0.0 - 13%

10 sets of duplicate samplings for Cs¹³⁷
avg. % deviation from mean: 4.1%, range 0.5 - 5.5%

5. DUPLICATE SAMPLING (cont'd)

B. Fallout (Monthly Pot Collections)

<u>HASL No.</u>	<u>dpm Sr⁹⁰</u> <u>per pot</u>
P5396	15.6
P5397	14.6
P5435	20.0
P5436	20.2
P5465	9.0
P5466	9.1
P5494	6.4
P5495	5.9
P5527	20.7
P5528	20.7
P5560	30.1
P5561	30.0
P5596	22.8
P5597	21.7
P5629	50.6
P5630	50.2
P5680	22.8
P5681	20.4
P5722	32.0
P5723	32.8
P5759	16.1
P5760	17.0

11 sets of duplicate samplings

Avg. % deviation from mean: 3.4%, range 0 - 11%

1
STABLE LEAD CONCENTRATIONS IN NEW YORK CITY FOODSTUFFS

D. C. Bogen (HASL)

ABSTRACT

Nineteen food categories which constitute the typical American diet have been analyzed for stable lead. Three complete diet samplings are reported. The dietary intake of lead obtained from the first study was about 285 $\mu\text{g/day}$, while the intake from the other two studies was between 70-80 $\mu\text{g/day}$. The higher results obtained for the first study are suspect because of possible contamination problems.

INTRODUCTION

The National Research Council's Committee on Biological Effects of Atmospheric Pollutants recommended⁽¹⁾ more extensive monitoring of lead in common food items, drinking water, and miscellaneous beverages. The committee's report concluded that the average American consumes more lead in food and beverages than he inhales from the air. Our laboratory started a modest program in 1966 to document the stable lead concentrations in the New York City diet. The food categories chosen for analysis were the same as in the HASL Tri-City diet program⁽²⁾ since these categories constitute a typical American diet. We have completed three diet samplings and each covers a specific time period when the food was purchased and analyzed. These time periods are (a) Fall 1966-Winter

1967, (b) Fall 1968-Winter 1969, and (c) Fall 1970-Winter 1971.

ANALYSIS

- A. The first diet sampling was analyzed by a method previously reported⁽³⁾.
- B. The method was modified for the second diet sampling. Samples were freeze-dried and then ashed in a low temperature asher (Tracerlab Model LTA 600). The ashed residues were dissolved in 1M HCl and the lead was extracted into methyl-isopropyl ketone⁽⁴⁾ as the iodide complex using Pb-212 tracer to monitor for any losses. Then the organic lead extract was measured directly on an atomic absorption spectrophotometer and the lead values obtained from a calibration curve.
- C. The method of analysis was slightly modified for the third diet sampling. The food samples were wet ashed with nitric acid after freeze-drying. The lead was isolated, purified, and measured as outlined in the second diet sampling.

RESULTS AND DISCUSSION

The lead results for the three diet samplings are reported in Tables I, II, and III. The yearly food intake values for each category were obtained from U. S. Department of

Agriculture statistics as summarized by Rivera⁽⁵⁾. The results for the first diet survey (1966-1967) indicate an average daily intake of about 285 μg . The results for the second and third diet surveys (1968-1969, 1970-1971) are in fair agreement with each other indicating an average daily intake of 70-80 μg . The lead content within a food category can vary by a factor of two from one sampling to another. We believe that the higher lead values obtained in the first diet study may be suspect because of contamination. In the first experiment the samples were prepared for analysis by wet ashing without first freeze-drying the samples. This made it necessary to use large quantities of HNO_3 and H_2O_2 to solubilize the sample and these reagents contain trace amounts of lead. The blank levels were 2-5 $\mu\text{g Pb}/\ell$ for HNO_3 and 50-200 $\mu\text{g}/\ell$ for H_2O_2 . Therefore, there is a significant uncertainty when the data were corrected for reagent contamination. In the last sampling the use of HNO_3 was restricted and no H_2O_2 was used so that contamination was kept to a minimum.

It is evident that our dietary lead results are lower than those of other investigators^(6,7). A major problem in any trace metal analysis is contamination and this may represent

the differences between our results and the other investigators. It should be noted that we have used Pb-212 tracer to monitor for any chemical losses in the procedure and that we employ large sample sizes (> 1 kg) so that sensitivity is not a problem.

Certainly these differences will be studied further, since there is the implication that ambient lead air concentration in urban environments may contribute as much to the lead intake as food. Measurements performed at our laboratory indicate an average lead air concentration of about $2 \mu\text{g}/\text{m}^3$ for New York City. Assuming a total daily inhalation volume of 20 m^3 the lead air intake would be about $40 \mu\text{g Pb/day}$ which is comparable to the dietary intake reported in the second and third food samplings.

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Table I
STABLE LEAD IN TOTAL DIET
 1966-1967 Sampling

<u>Category</u>	<u>Yearly Intake (kg)</u>	$\mu\text{g Pb}$ <u>g wet wt</u>	<u>Total $\mu\text{g Pb}$ yr</u>
Fresh Fish	8	0.16	1280
Shellfish	1	0.31	310
Poultry	20	0.30	6000
Meat	79	0.42	33180
Eggs	15	0.22	3300
Fresh Fruit	59	0.072	4250
Canned Fruit	11	0.25	2750
Fresh Vegetables	48	0.12	5760
Canned Vegetables	22	0.44	9680
Root Vegetables	10	0.068	680
Potatoes	38	0.17	6460
Macaroni	3	0.077	230
Rice	3	0.044	130
Juice	28	0.091	2550
Dry Beans	3	0.024	70
Flour	34	0.041	1390
Bakery Products	44	0.39	17160
Whole Grain Products	11	0.13	1430
Milk	200	0.038	7600

Table IISTABLE LEAD IN TOTAL DIET

1968-1969 Sampling

<u>Category</u>	<u>Yearly Intake (kg)</u>	$\mu\text{g Pb}$ <u>g wet wt</u>	<u>Total $\mu\text{g Pb}$ yr</u>
Fresh Fish	8	0.049	390
Shellfish	1	N.A.	-
Poultry	20	0.042	840
Meat	79	0.060	4740
Eggs	15	0.045	680
Fresh Fruit	59	0.063	3720
Canned Fruit	11	0.17	1870
Fresh Vegetables	48	0.059	2830
Canned Vegetables	22	0.033	730
Root Vegetables	10	0.039	390
Potatoes	38	0.089	3380
Macaroni	3	N.A.	-
Rice	3	0.020	60
Juice	28	N.A.	-
Dry Beans	3	0.049	150
Flour	34	0.022	750
Bakery Products	44	N.A.	-
Whole Grain Products	11	0.050	550
Milk	200	0.016	3200

N.A.: Not analyzed.

Table III
STABLE LEAD IN TOTAL DIET
1970-1971 Sampling

<u>Category</u>	<u>Yearly Intake (kg)</u>	<u>μg Pb g wet wt</u>	<u>Total μg Pb yr</u>
Fresh Fish	8	0.057	460
Shellfish	1	0.19	190
Poultry	20	0.068	1360
Meat	79	0.038	3000
Eggs	15	0.052	780
Fresh Fruit	59	0.041	2420
Canned Fruit	11	0.19	2090
Fresh Vegetables	48	0.035	1680
Canned Vegetables	22	0.060	1320
Root Vegetables	10	0.041	410
Potatoes	38	0.051	1940
Macaroni	3	0.052	160
Rice	3	0.11	330
Juice	28	0.086	2410
Dry Beans	3	0.11	330
Flour	34	0.032	1090
Bakery Products	44	0.10	4400
Whole Grain Products	11	0.083	910
Milk	200	0.021	4200

✓ Comparison of the Original and Modified
High-Volume-3000 Air Ejector Samplers

by

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Abstract

A modification of the High-Volume-3000 air ejector sampler (HV3K) has been developed and flight tested against an original HV3K sampler. Results indicated the original sampler became contaminated with radioactive debris during its ascent to sampling altitude. This confirmed the earlier results reported by Ashenfelter and Telegadas (1970). The modified HV3K sampler is considered to be contamination-free during its ascent to sampling altitude.

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Introduction

The U.S. Atomic Energy Commission balloon-borne atmospheric sampling program, Projects Cold Ash and Hibal, (Krey and Kleinman, 1970) had employed a High Volume-3000 Sampler (HV3K) to collect radioactive samples at an altitude of 41 km. Ashenfelter and Telegadas (1970) questioned the integrity of this sampler. They demonstrated that during the sampler's ascent to collection altitude, air was forced down the diffuser tube, through the filter paper and out the inlet door, if the inlet door was partially open. This would lead to contamination of the filter paper (IPC-1478) with fission products which were and still are more abundant at lower altitudes. These samplers were taken out of operation as of October 1969 and the scientific community alerted to the questionableness of past data based on the HV3K sampler collections (Krey and Kleinman, 1970).

A modification of the original HV3K samplers was made by Ashenfelter by redesigning the sampler doors to make them air tight. This report describes the modified sampler and gives the results of the intercomparison test of the original and modified sampler.

Description of the Modified Sampler

A schematic diagram of the modified sampler is shown in Figure 1. This sampler is the same as the original sampler reported by Ashenfelter and Telegadas (1970) with two exceptions. The inlet and outlet doors were redesigned and a pressure relief port installed into the body of the sampling module.

In the original sampler, the inlet and outlet doors were spring loaded and operated independently of one another. During ascent of the module, the outlet door was fastened in an open position and the inlet door was fastened in a closed position. In the original design, the outlet door could only be actuated in one direction whereas the inlet door could be actuated in both directions. At about 29 km, the inlet door was opened by a firing mechanism as the system continued to ascend to float altitude of about 41 km. At sampling altitude air was pumped through the filter media for a predetermined time. Upon termination of sampling, the inlet and outlet doors were closed and the system descended by parachute.

It was observed that when the inlet door was secured in its closed position, it would not, in all instances, make a proper seal with the sampling module body (Ashenfelter and Telegadas 1970). Whenever this situation existed, a small fraction of the air intercepted by the diffuser tube during ascent would be forced through the sampling module by ram air pressure (ascent velocity $\sim 5 \text{ m sec}^{-1}$). If the sampler system passed through a relatively high level of radioactivity, the

filter paper would become contaminated, such that fictitiously high concentrations of fission products would be reported as having been sampled at float altitude.

The sampling doors were redesigned and connected to pneumatic actuators and are now in a closed position during ascent. At sampling altitude the high pressure air ejector pump gas supply is turned on which operates the actuators to open both doors simultaneously and also functions the air ejector to pull air through the filter paper. At termination of sampling, the gas supply is turned off and a tight closure of the inlet and outlet doors is assured by the return spring within the pneumatic actuator. The package then descends by parachute.

During ascent and descent, the sampling module doors are in an air tight configuration. To equalize pressure between the inside of the module and the ambient air, a pressure relief port was installed. A hole, 5 cm in diameter, was cut into the body of the sampling module and a screened plug containing 3 layers of IPC-1478 filter paper was inserted.

Comparison of the Original and Modified Samplers

On December 12, 1970, an intercomparison test involving the original and modified HV3K units was undertaken at San Angelo, Texas (31°N). The experimental sampling configuration was similar to that shown in Figure 1 with several exceptions. First, the gas supply was not flown since no sampling would take place at float altitude. Second,

an original HV3K sampler was placed with its diffuser, also in a vertical position, directly below the modified sampler shown in Figure 1. In anticipation of low flow rates during ascent, the two PR-3 60 degree blade flowmeters were replaced by more sensitive PR-3 30 degree blade flowmeters. The flow rate through the filter media could be obtained by both on-board recording and telemetry of flowmeter rotational speed. On a single flight therefore, it was planned to have both sampling modules ascend to an altitude of 30 km and then descend.

The rigging crew of Detachment 31, 6th Weather Wing, U.S. Air Force at Goodfellow AFB, San Angelo, Texas, set up the experimental flight under the direction of T. Ashenfelter. The original HV3K module (Sampler A) had the outlet door fastened in the "open" position and the inlet door in the "closed" position, simulating past operational conditions. The modified HV3K module (Sampler B) had both sampling doors in a closed position.

The complete system ascended to about 30 km whereupon the outlet door of Sampler A was closed and the sampling modules descended by parachute. As the balloon ascended, the telemetry of flowmeter speed was monitored for both modules. Telemetry was received from Sampler A, indicating very low flow while telemetry from Sampler B indicated zero flow. Upon recovery of the package, the on-board flowmeter recorder from Sampler A indicated a rotational speed of about 8 cycles per second (cps) to about 7 km, this is equivalent to about 0.6 ambient

cubic meter of air per minute (ACM/min) passing through the filter media (Wood, et al., 1965; Loysen, 1966). The rotational speed decreased to about 6 cps at an altitude of 13 km, equivalent to about 0.5 ACM/min. Above 13 km the rotational speeds were very low and the recorder chart could not be read accurately. As will be shown, the highest concentrations of fission products in the atmosphere at about 30°N existed between 13 to 23 km. If one assumes that the rotational speed of the flowmeter between 13-23 km was between 4 to 2 cps, for the experimental flight of December 12, 1970 a volume of air of about 11 to 6 ACM is estimated to have passed through the filter paper in this altitude range. It will be shown that this volume of air is in fair agreement with the radioactivity results presented in Table 1. The on-board flowmeter recorder for Sampler B indicated zero flow through the filter media.

Radioactivity Results from the Experimental Flight

The filter papers from Sampler A and B were subjected to radiochemical analysis to see if any contamination occurred during ascent. The Sr-89 and Sr-90 data (pCi/total paper) for the December 12, 1970 experimental flight are shown in Table 1. Sampler A, in which the outlet door was in an open position and in which the flowmeter recorder indicated air flow through the filter media showed considerable Sr-89 activity on its filter paper whereas Sampler B, in which the sampling doors were in a closed position indicated the Sr-89 value to be below

the limits of detection. The low Sr-90 values found on both filter papers is considered to be background Sr-90 indigenous to the 8-square-foot filter paper (Krey, 1971).

The concentration profile (picocuries per ambient cubic meters of air, pCi/ACM) of Sr-89 and Sr-90 based on aircraft and balloon flights (Krey and Kleinman, 1971) to 27 km is shown in Figure 2. It is assumed that this profile which was based on observations between December 23, 1970 and February 24, 1971 prevailed over the balloon launch site of San Angelo, Texas on the day of the experimental flight, December 12, 1970. As is quite evident, the highest concentrations occurred from 13 to 23 km. The Sr-89 concentrations at and above 23 km were below the limits of detection whereas the Sr-90 concentration at 27 km was 3×10^{-4} pCi/ACM (beyond the limits of the graph), a factor of about 50 less than the concentration at 23 km.

The HV3K diffuser opening has a diameter of about 0.3 meter and during ascent between 13 to 23 km this diffuser would sample a column of air of about 700 ambient cubic meters if there were no obstruction to free flow. Since the outlet door of the original HV3K sampler was in an open position during ascent, the main obstructions to free flow would be the filter media and the inlet door. If the inlet door was partially open, one would then expect a small fraction of this air column to pass through the filter media.

The average concentration of Sr-89 and Sr-90 between 13-23 km based on the concentration profiles found in Figure 2 are shown in

Table 1. Also given in Table 1 is the ambient cubic meters of air (ACM) required to pass through the filter paper during ascent from 13 to 23 km to account for the activity found on the filter paper. As can be seen, a leakage of 4.2 ACM of air would be needed to account for the Sr-89 value found on the filter paper of Sampler A. This volume of air is 0.6% of the volume of air swept out by the HV3K diffuser if there were not obstructions to free flow. The volume, computed from the flowmeter (11-6 ACM of air) mentioned earlier, is in fair agreement with the leakage (based on the Sr-89 activity) as are the leakage results found on previous experimental flights of the original HV3K reported by Ashenfelter and Telegadas (1970).

The average concentration of Sr-90 from 13 to 23 km from the profile in Figure 2 is 0.05 pCi/ACM. A leakage of 4 ACM of air would account for 0.2 pCi of the 1.2 pCi/total filter paper found. This relatively small contamination is within the range of Sr-90 contamination found on blank HV3K filter papers (Krey, 1971).

For Sampler B where both doors were in a closed position during ascent and descent and the flowmeter indicated zero flow through the sampling module, the Sr-89 activity found on the filter paper was below the limits of detection. Since the limit of detection is about 2 pCi/total paper (Krey, 1971), if air did pass through this system, it would have to be at most about 1 ACM of air which is a factor of 4 less than assumed to have leaked through Sampler A.

It would appear from this experimental flight that if any leakage occurred in the modified sampler it is of no significance.

Preliminary Results from Operational Flights of the Modified HV3K Sampler

The modified HV3K Sampler has been flown operationally at about 41 km over Mildura, Australia and San Angelo, Texas since its inter-comparison with the original HV3K Sampler in December 1970. The preliminary radioactivity results of these two flights are shown in Table 2. Strontium-89 was not detected in resolvable amounts on either flight. Over Mildura, balloon sampling at 24 and 27 km by a Direct Flow Sampler and at 33 and 36 km by an Air Ejector Sampler in March-April 1971 indicated Sr-89 concentrations to be below the limits of detection (Krey and Kleinman, 1971). Aircraft and balloon data indicated a layer of relatively high concentrations between 10 to 21 km during March 1971. It would appear that the modified HV3K sampler which sampled 25 standard cubic meters of air (SCM) at 41 km did not leak while ascending to the sampling altitude. The Sr-90 activity found on the Mildura flight can be compared with the activity found on 8 square feet of blank filter paper. Results indicate the Sr-90 activity to be within the background contamination level of the filter media.

Balloon flights over San Angelo, Texas between 27 to 37 km in May-June 1971 indicate Zr-95 concentrations to be below the limits of detection (no Sr-89 data available at this time). Aircraft data are not available to estimate the layer of highest concentrations. From the balloon data below the sampling altitude of the modified HV3K sampler, it appears that the modified sampler did not leak as it

ascended to 41 km. The Sr-90 activity found on the June 1, 1971 flight also appears to be within the limits of the background contamination of the filter media.

Summary

The integrity of the original HV3K sampler used in collection of radioactivity samples at 41 km was questioned. It was demonstrated that during ascent, air may be forced down the diffuser tube, through the filter paper and out the inlet door, if the inlet door was partially open (Ashenfelter and Telegadas, 1970). A modification to the sampling doors has been developed by the Air Resources Laboratories, NOAA. This modification together with a modification of the sampling module (pressure relief port) to insure a contamination-free system has been flight tested against an original HV3K sampler. Results from this test, together with operational flight results, indicate that the filter media of the modified HV3K sampler does not appear to become contaminated by radioactive debris during its ascent to sampling altitude.

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Table 1. HV3K Comparison Test of December 12, 1970
(All data decay corrected to Dec. 12, 1970)

Sampler			<u>13-23 km</u>		<u>Leakage Required</u>	
	Sr-89	Sr-90	Sr-89	Sr-90	Sr-89	Sr-89
	(pCi/total paper)		(pCi/ACM)		(ACM)	(%)
A (original)	9.2	1.2	2.2	0.05	4.2	0.6
B (modified)	ND	1.2	2.2	0.05	-	-

pCi/total paper: Picocuries found on total paper by
radiochemical analyses.

pCi/ACM: Picocuries per ambient cubic meter of air; average
concentration between 13-23 km from fig. 2.

$$\text{Leakage (ACM)} = \frac{\text{pCi/total paper}}{\text{pCi/ACM}}$$

$$\% = \frac{\text{Leakage (ACM)}}{700 \text{ ACM Swept out by diffuser}} \times 100$$

ND: Isotope was not detected in resolvable amounts.

Table 2. Radioactivity Data from Operational
Flights of the Modified HV3K Sampler

Date	Alt (km)	Volume Air Sampled (SCM)	Sr-89 pCi/total	Sr-90 paper
Mildura, Australia, 34°S March 18, 1971		25	ND	0.8
San Angelo, Texas, 31°N June 1, 1971		29	ND	1.4
Blank (1)		--	ND	1.6

(1) 8 square feet of Blank IPC-1478 filter paper

ND Isotope was not detected in resolvable amounts.

Fig.1: Schematic Diagram of Modified HV3K Air Ejector Filter Sampler

✓ Carbon-14 Measurements in the Stratosphere
From a Balloon-Borne Molecular Sieve Sampler

by

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Abstract

A new sampler for collection of atmospheric CO₂ utilizing molecular sieve adsorption has been flown on operational USAEC balloon flights beginning in mid-1970 at four locations in the northern and two locations in the southern hemisphere at altitudes ranging from 21 to 37 km. The CO₂ collections were analyzed radiometrically for excess carbon-14 (C-14 due to nuclear weapons testing) and by mass spectrometric analysis for δ C-13 and δ O-18. Only the excess C-14 and δ C-13 data are given. Although the δ O-18 data are not presented, a discussion of the results is given.

The δ C-13 values decrease with altitude from 21 to 37 km. This decrease is attributed to fractionation in the molecular sieve beds during the collection of CO₂ rather than in nature or in the laboratory.

The excess C-14 data are presented in two latitudinal cross-sections from which the stratospheric excess C-14 burden was determined.

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Carbon-14 Measurements in the Stratosphere
From a Balloon-Borne Molecular Sieve Sampler

I. Introduction

The U. S. Government has conducted an extensive atmospheric program of whole air collection in the troposphere and stratosphere since 1953. These atmospheric samples were analyzed for C-14 collected in the form of CO_2 . Excess C-14 (C-14 due to nuclear testing) data have been derived and the results made available (Hagemann, et al., 1959, 1965; ESSA, 1966, 1967, 1969; NOAA, 1971). Recently, these data were used to determine the seasonal atmospheric distribution and inventories of excess C-14 from March 1955 to July 1969 (Telegadas, 1971a). The observed decreases of the excess C-14 inventory in the troposphere and stratosphere has been used by Machta (1971) to estimate the exchange of carbon dioxide between the atmosphere and the biosphere and oceans.

The high altitude balloon sampling portion (20 to 30 km) of the whole air collection program terminated in July 1966 while the aircraft program, which sampled in the troposphere and lower stratosphere to 20 km, continued until its termination in July 1969. The balloon portion of the C-14 program was operated for a year beginning in mid-1970 using a molecular sieve collector designed to replace the whole air balloon samplers. Plans are being discussed which may lead to a re-establishment of a somewhat modified version of the aircraft whole air sampling program supplemented by a continuation of the balloon molecular sieve sampler program.

The molecular sieve sampler has been flown experimentally on the operational USAEC High Altitude Balloon Sampling Program (Krey and Kleinmann, 1971) on flight conducted over San Angelo, Texas (31°N) at altitudes ranging from 21 to 37 km. The experimental data which were collected from mid-1968 to early 1970 together with a description of the sampler and laboratory techniques for determining the excess C-14 specific activity have been reported by Ashenfelter, et al., (1972).

For a year beginning in mid-1970, the molecular sieve samplers were flown operationally on the USAEC balloon flights at four locations in the northern hemisphere and two locations in the southern hemisphere. This paper repeats, where necessary, portions of the Ashenfelter, et al., (1972) report together with the data collected through mid-1971.

Future results of the C-14 molecular sieve program will be published as they become available in the USAEC Health and Safety Quarterly Summary Reports.

II. Description of the Carbon-14 Molecular Sieve Sampler

The balloon-borne C-14 molecular sieve sampler which was designed to collect CO₂ at altitudes ranging from 20 to 37 km is shown in figure 1. The complete system, containing Linde Type 4A molecular sieve (1/8 inch diameter adsorbent pellets), helium primary gas supply and necessary hardware, weighs about 30 kg. The air ejector principle is used to move ambient air at a known flow rate through the two collection beds.

The C-14 sampler was tested in the High Altitude Test Chamber of the USAEC Health and Safety Laboratory in New York City to determine the proper air ejector design and operating parameters to insure that a sufficient volume of CO₂ would be collected with approximately 100% efficiency at selected altitudes. Experimental flights over San Angelo, Texas, were conducted from mid-1968 to mid-1970 to verify the chamber results (Ashenfelter, et al., 1972).

III. Sampling Locations

The new C-14 sampler can readily be flown on the USAEC balloon-borne particulate sampling missions. Both samplers (molecular sieve and particulate sampler) were normally flown together at altitudes of 21, 24, 27, 32, and 37 km at the following locations:

<u>Location</u>	<u>Latitude</u>
Eielson Air Force Base, Alaska	65°N
Sioux City, Iowa	42°N
San Angelo, Texas	31°N
Albrook Air Force Base, Panama Canal Zone	9°N
Longreach Queensland, Australia	23°S
Mildura, Australia	34°S

The complete sampling system is lifted to a pre-determined float altitude where sampling begins for a pre-determined time. Upon completion of sampling, the system descends by parachute and is recovered.

The molecular sieve adsorbents are returned to Argonne National Laboratory (ANL) for quantitative determination of adsorbed CO_2 and for C-14, C-13 and O-18 analyses.

IV. Determination of Excess Carbon-14

The techniques for separating CO_2 from the molecular sieve pellets and the C-14 counting techniques are given by Ashenfelter, et al. (1972). In addition, a small aliquot of CO_2 is routinely taken for C-13 and O-18 mass spectrometric analysis.

In the determination of excess C-14, a known quantity of purified CO_2 is transferred into one of six 2-liter low-background counters. The data from these counters are reported in "absolute" disintegrations per minute per gram of carbon (dpm/gC). The technique employed by carbon dating laboratories is to avoid absolute calibration by taken the ratio of the unknown sample to a National Bureau of Standard (NBS) oxalic acid C-14 standard converted to CO_2 . The actual value of pre-bomb CO_2 (modern wood) is considered to be equal to 0.95 NBS oxalic acid standard or 13.1 dpm/gC.

The "absolute" activity of the ANL standard gas (2000 dpm/l) has been used to calibrate all counters and the NBS oxalic acid standard. The ANL counters gave a mean value for 0.95 oxalic acid standard of 13.17 ± 0.04 dpm/gC at the 2-sigma confidence level. Thus, ANL confirms within experimental error the generally accepted value of 13.1 dpm/gC for the oxalic acid standard.

The numbers reported in this paper as excess specific activity (Table 1) are the measured dpm/gC minus 13.1 dpm/gC. In relating excess activity to bomb C-14 concentrations (atoms per gram of air) we have used 74×10^5 atoms/g of air as the pre-bomb background (Hagemann, et al., 1965) assuming a CO₂ concentration in air of 313 ppm (by volume).

V. Stratospheric Carbon-14 Measurements

The experimental and operational data using the molecular sieve sampler are given in Table 1. An explanation of the table is given:

Column 1. Flight No.

The experimental flights conducted over San Angelo, Texas are designated by an X.

Column 2. Date

The date of collection is given by three two digit groups of month, day and year.

Column 3. Alt.

The mean pressure altitude of the collection is given in kilometers.

Column 4. Bed No.

The data from the two sampling beds are listed as Bed 1 (the primary sampling bed) and Bed 2 (the back-up bed to detect any breakthrough of CO₂ from Bed 1).

Column 5. Vol. CO₂.

The volume of CO₂ in cubic centimeters (cc) collected at STP on each bed.

Column 6. δ C-13

The measured C-13/C-12 value expressed as the deviation from Craig's (1957) limestone standard in units of per mil (0/00).

$$\delta \text{ C-13} = \left[\frac{\text{C-13/C-12 (Sample)}}{\text{C-13/C-12 (P.D.B. Standard)}} - 1 \right] \times 1000$$

The standard error of the determination is estimated at 0.2%.

Column 7. Excess Spec. Act.

These numbers are the observed specific activity (dpm/gC) minus 13.1 dpm/gC. This is therefore the excess specific activity due to nuclear weapons testing.

The counting error of the observed specific activity at the 95% confidence level is less than 5% in all samples and less than 3% in 90% of the samples.

Column 8. C-14 Excess.

These numbers are the excess specific activity (dpm/gC) converted to C-14 excess concentration in units of 10^5 atoms per gram of air.

$$\text{C-14 Excess} = \left[\text{Excess Spec. Act. (dpm/gC)} \right] \left[\frac{74 \times 10^5 \text{ atoms/g air}}{13.1 \text{ dpm/gC}} \right]$$

Column 9. C-14 Excess, Wt. Avg.

These values are the weighted average of the excess C-14 determination from Bed 1 and 2 weighted by the volume of

CO₂ collected on each bed and is defined as:

$$\text{C-14 Excess (Wt. Avg.)} = \frac{(\text{C-14 Excess})_1 (\text{Vol CO}_2)_1 + (\text{C-14 Excess})_2 (\text{Vol CO}_2)_2}{(\text{Vol CO}_2)_1 + (\text{Vol CO}_2)_2}$$

Column 10. Remarks

The meaning of each remark is given at the bottom of each page of Table 1.

VI. Distribution of δ C-13 with Altitude

As mentioned previously, aliquots of CO₂ are taken for the measurement of the C-13/C-12 ratio. Knowing this ratio, a correction can be applied to the C-14 counting rate to correct for any fractionation, relative to a standard, of the carbon isotopes (Broecker and Olson, 1961). This fractionation may have taken place in nature, during collection of CO₂ or in the laboratory.

The δ C-13 of uncontaminated air is assumed to be -7.1 0/00 as suggested by Craig (1953) and Keeling (1961) from ground level air. About 100 whole air samples collected over San Angelo, Texas (31°N) between 20 and 33 km from January 1964 to May 1966 had an average CO₂ concentration of 308 ± 5 ppm by volume (Hagemann, et al., 1965; ESSA, 1966, 1967, 1969). These samples were analyzed by ANL and yielded an average δ C-13 value of -8.1 0/00 with a standard deviation about the mean of 1.0. The average δ C-13 value, -8.1 0/00, is in agreement with the -7.1 0/00 reported by Craig and Keeling.

Young (1967, 1971) used Linde type 4A molecular sieve for the adsorption of CO₂ from the air at ground level and at aircraft

altitudes. Aliquots of uncontaminated CO_2 from 25 atmospheric samples yielded an average $\delta^{13}\text{C}$ of -9.2 ‰ with a range of -10.3 ‰ to -7.7 ‰. The average $\delta^{13}\text{C}$ and its standard deviation was determined for the molecular sieve data listed in Table 1. The results are shown in Table 2 for Bed 1 (Primary), Bed 2 (Breakthrough) and the weighted average for both beds (weighted by CO_2 collected) together with the results from the whole air collections mentioned previously. The results indicate that the scatter about the mean value for the molecular sieve $\delta^{13}\text{C}$ data (Bed 1 and 2 are weighted for CO_2 content) is greater than that of the whole air collector. The $\delta^{13}\text{C}$ mean value for Bed 1 (where, in nearly all cases, most of the CO_2 was adsorbed) is in agreement with the mean value of the whole air collector. Table 2 shows a significant difference between the average $\delta^{13}\text{C}$ value of Bed 1 and Bed 2 indicating an enrichment of ^{13}C molecules in Bed 2 relative to Bed 1.

In exploring the fractionation effect further, the $\delta^{13}\text{C}$ values determined from both the molecular sieve and whole air collectors were plotted as a function of collection altitude. This distribution is shown in figure 2 for Bed 1 and Bed 2 while the weighted average of Bed 1 and Bed 2 together with the whole air collector are shown in figure 3. The equation for the "line of best fit" together with the standard deviation about that line is shown on both figures. A statistical analysis indicates that the level of significance is 0.02 for Bed 1 and 0.01 for Bed 2 signifying the decrease of $\delta^{13}\text{C}$ with altitude did not happen by chance. All individual stations showed about the same trend of $\delta^{13}\text{C}$

with altitude evident in the mean pictures of figures 2 and 3.

The $\delta^{13}\text{C}$ -13 data from the whole air collector (figure 3) were determined by ANL. Since this distribution does not show any altitude dependency in the $\delta^{13}\text{C}$ -13 results, it is apparent that fractionation in the molecular sieve system (also analyzed by ANL) took place during the collection of CO_2 and had not taken place in nature or in the laboratory.

To explore this isotopic fractionation with altitude, laboratory experiments are being conducted to simulate altitude, bed temperature and flow rates as well as an examination of these parameters from the actual flight records to see if an explanation can be found.

VII. Stratospheric Distribution of $\delta^{18}\text{O}$

Aliquots of CO_2 adsorbed by the molecular sieve material were also taken for the measurement of the oxygen isotopic composition of atmospheric CO_2 . The definition of $\delta^{18}\text{O}$ -18 (Hagemann, et al., 1965) is analogous to that for $\delta^{13}\text{C}$ -13:

$$\delta^{18}\text{O}-18 = \left[\frac{0-18/0-16 \text{ (Sample)}}{0-18/0-16 \text{ (P.D.B. Standard)}} - 1 \right] \times 1000$$

Craig and Begemann (1970) using the $\delta^{18}\text{O}$ -18 data reported by Hagemann, et al. (1965) for the period 1953-1964 indicated the average stratospheric value to be -5.0 ‰. They showed that collected data from the whole air sampler,

"... do not reflect the actual isotopic composition of oxygen in CO_2 but rather an artifact of the sampling technique."

We have calculated the mean $\delta^{18}O$ and standard deviation about the mean for the whole air samples collected between 20 to 33 km over San Angelo, Texas (31°N) between January 1964 to May 1966. (Hagemann, et al., 1965; ESSA 1966, 1967, 1969). An average value of -4.9 ± 5.2 ‰ was determined which was not dependent on altitude. This is in agreement with the value, -5.0 ‰ reported by Craig and Begemann.

The $\delta^{18}O$ determined from the molecular sieve samples collected between 20 to 37 km had a mean and standard deviation of -18.1 ± 4.1 ‰, and was also found to be independent of collection altitude.

During a recent meeting held on August 24, 1971 at ANL to discuss the collection of atmospheric gases using molecular sieve material, H. Gote Ostlund stated that,

"... there was oxygen isotopic exchange in both influent H_2O and CO_2 with the SiO_2 and/or residual water in the sieve so that isotopic oxygen analysis with molecular sieves is "impossible" (Krey 1971).

Even though the $\delta^{18}O$ of atmospheric CO_2 desorbed from the molecular sieve material was determined, it is not reported here due to the findings of Craig and Begemann (1970) and the statement by Ostlund (Krey 1971). They are however available from the authors.

VIII. Latitudinal Distribution of Stratospheric Excess C-14

The data listed in Table 1 permits a reasonable inventory of the excess C-14 in the stratosphere above 20 km and its distribution for the first time since early 1959 when four balloon stations were in

operation (Telegadas, 1971). After 1959, only one balloon station was used in the collection of whole air samples (San Angelo, Texas, 31°N) and this station terminated in July 1966. The latitudinal distribution of excess C-14 as a function of altitude for two periods, September - December 1970 and January - June 1971, are shown in figures 4 and 5. The northern and southern hemisphere excess C-14 burden between 21 and 37 km (Table 3) was estimated using the distribution in these figures.

Telegadas (1971) computed the stratospheric burden of excess C-14 in the northern and southern hemispheres between 21 and 31 km from similar cross sections from June 1964 to July 1966 with only one sampling location. The effective half residence time of excess C-14 for the whole hemispheric compartment from 21 to 31 km is estimated to be about 60 months. Extrapolating the mid-1964 to mid-1966 excess C-14 inventory with a 60-month half residence time, assuming no new inputs into this compartment by large yield nuclear tests, one finds an estimated inventory for the same periods as given in figures 4 and 5 of 16 and 15×10^{26} excess C-14 atoms. These extrapolations are listed in Table 3 where they can be shown to be 40% less than the observed inventories. If this comparison is valid, about 40% of the high altitude excess C-14 can be attributed to the Chinese and French high yield tests conducted between mid-1967 to mid-1970. This conclusion must be tempered by the large uncertainty in the inventories between mid-1964 to mid-1966 since they are based on a limited number of samples at only one location.

An examination of the data analyzed from the USAEC particulate samplers flown on the same balloon with the molecular sieve sampler reveals that the short-lived fission product Zr-95 (65 day half-life) and the long-lived Cs-137 (30 year half-life) in balloon samples collected above 20 km are one to two orders of magnitude less than samples collected at and below 21 km by balloons and aircraft. Based on fission product considerations, it is tempting to speculate that virtually no gaseous C-14 was injected above 21 km and the maximum concentrations of excess C-14 observed at about 25 km in both hemispheres (figures 4 and 5) are due possibly to the nuclear tests conducted by the USSR and U.S. in 1961-1962.

Important information about the motions of the atmosphere can be obtained from the investigation of the distribution of radioactive substances in the atmosphere and their changes in time. Unique tracers such as W-185, Rh-102, Cd-109, Pu-238 (Frankel and Salter, 1961; Telegadas, 1971b; Telegadas and List, 1964; 1969) and the activity ratios of fission products from nuclear tests (Feely and Spar, 1960; Gudiksen, et al., 1968; Kalkstein, 1962; List and Telegadas, 1969; Machta, et al., 1970) have proven very useful in yielding information on the transport and diffusion processes within the stratosphere. Before any interpretation of a distribution is attempted, one should try to determine where and when the tracer material started from.

The latitudinal distribution of excess C-14 in figures 4 and 5 indicate maximum concentrations in temperate and polar latitudes of

both hemispheres at about 25 km and a minimum concentration over the equatorial region. This pattern may be compared with fission product distributions of Zr-95 and Cs-137 based on aircraft data to about 20 km and balloon data above for November 1970 and February 1971 as given by Krey and Krajewski (1971). The maximum concentrations of these fission products in the northern hemisphere temperate and polar regions lie between 14 to 18 km and are attributed primarily to the Chinese high yield nuclear test conducted at 40°N in October 1970. The southern hemisphere maximum is found between 15-19 km in the equatorial and temperate latitude region (no sampling beyond 50°S) and is ascribed to the two high yield French nuclear tests conducted at 22°S between May to August 1970. The maximum concentration in both hemispheres in general slope along surfaces oriented downward toward the poles. It is significant that the fission-product maximum is 5 to 10 km lower in altitude than the excess C-14 distributions shown in figures 4 and 5. Telegadas and List (1969) have shown that particle settling of radioactive debris to be insignificant after a few weeks or so of a nuclear test. It therefore seems unreasonable that the fission product maximum concentrations would be this much lower than the excess C-14 maximum concentrations if they were from the same events. It would appear that the maximum concentrations of excess C-14 observed at about 25 km in both hemispheres, figures 4 and 5, are due primarily to the high yield nuclear tests conducted prior to the Chinese and French high yield nuclear tests of 1967-1970.

It is difficult to interpret the observed distributions of excess C-14 shown in figures 4 and 5 in terms of atmospheric processes due to the uncertainties as to the source of excess C-14. A reactivation of the aircraft whole air sampling program and a continuation of the collection of CO₂ balloon samples would prove valuable in the investigation and refinement of the exchange rates of CO₂ between the atmosphere and the biosphere and oceans as has been recently reported by Machta (1971).

IX. Acknowledgements

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TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS

EIELSON AFB, ALASKA 65N

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS ⁵ (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
101	091970	20.9	1	3800	- 7.1				
			2	200	- 4.6				
102	091970	24.3	1	2575	- 8.0	{ 24.6	{ 138.7		
			2	185	- 1.2				
103	092070	31.0	1	740	- 7.5	21.3	120.1	120.1	A
			2	325	- 9.7				
104	092170	27.0	1	1175	-12.7	24.8	139.9	140.5	
			2	760	- 3.4	25.1	141.6		
105	060271	24.2	1	3800		20.7	116.7	117.1	
			2	955		21.0	118.4		
106	060371	31.8	1	800		23.3	131.4	131.2	
			2	675		23.2	130.8		
107	060471	27.4	1	2850		23.6	133.1	131.9	
			2	1560		23.0	129.7		

 A. BED 1 AND 2 COMBINED FOR SPECIFIC ACTIVITY ANALYSIS

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS
SIOUX CITY, IOWA 42N

FLIGHT NO.	DATE	ALT	BED NO.	VOL. CO-2	δ C-13 CONC.	EXCESS SPEC. ACT.	C-14 EXCESS	C-14 EXCESS WT. AVG.	R E M A R K S
		(KM)		(CC)	(PER MIL)	(DPM/GC)	⁵ (10 ATOMS/G AIR)		
13	100170	30.9	1	1180	- 8.1	23.7	133.7	133.7	
			2	700	- 8.2	23.7	133.7		
14	100370	24.2	1	3360	- 2.6	25.5	143.8	142.5	
			2	980	- 5.3	24.4	137.6		
15	100470	27.5	1	3200	- 5.3	24.4	137.6	137.6	
			2	1600		24.4	137.6		
16	101370	20.9	1	5230	-12.0	22.1	124.6	125.3	
			2	460	- 2.3	23.7	133.7		
17	101470	36.1	1	400	- 8.0	{ 22.2	{ 125.2	125.2	A
			2	400	- 9.3				
18	021571	21.3	1	5700	- 4.7	23.1	130.3	130.5	
			2	500	+ 3.7	23.4	132.0		
19	021771	24.6	1	3900	- 8.3	22.1	124.6	125.5	
			2	850	- 0.3	22.9	129.2		
20	022471	27.9	1	2870	- 7.2	24.2	136.5	133.7	
			2	1200	- 8.0	22.5	126.9		
22	040471	27.5	1	3400	- 9.8	24.3	137.1	134.4	
			2	1350	- 1.2	22.6	127.5		

A. BED 1 AND 2 COMBINED FOR SPECIFIC ACTIVITY ANALYSIS

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS
SIOUX CITY, IOWA 42N

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS 5 (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
23	040571	32.3	1	1050	-12.0	19.9	112.2	116.0	
			2	575	- 6.3	21.8	123.0		
24	040671	24.7	1	3400	- 1.9	20.5	115.6		
			2	1200	-----	-----	-----		A
25	040771	21.6	1	4700	- 9.1	21.9	123.5	122.9	
			2	1075	- 1.2	21.4	120.7		

A. LOST IN PURIFICATION

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS

SAN ANGELO, TEXAS 31N

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS 5 (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
1272X	052468	36.6	1	407	-12.2	28.0	157.9	155.6	
			2	368	-12.9	27.5	155.1		
1287X	110568	36.3	1	326	-12.2	24.4	137.6	147.0	
			2	247	- 4.5	28.3	159.6		
1293X	012169	29.2	1	2804	-14.1	27.5	155.1	154.9	
			2	473	- 5.3	27.3	154.0		
1296X	021169	29.8	1	977	-14.7	24.9	140.4		
			2	99	-22.3	-----	-----		A
1301X	031369	20.8	1	4120	- 7.2	28.1	158.5	158.4	
			2	679	-----	28.0	157.9		B
1302X	032169	29.5	1	2163	- 8.9	28.0	157.9	157.2	
			2	716	- 2.6	27.5	155.1		
1305X	051369	36.4	1	1795	- 4.0	10.1	57.0	60.7	
			2	2168	- 6.3	11.3	63.7		
1306X	051869	32.8	1	1086	- 9.6	27.9	157.4	154.1	
			2	518	- 6.8	26.1	147.2		
1309X	070169	24.3	1	4765	- 9.0	25.3	142.7	141.5	
			2	1100	-----	24.2	136.5		B

A. SPECIFIC ACTIVITY NOT COUNTED FOR BED 2

B. NO ANALYSIS OF δ C-13 FOR BED 2

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS
SAN ANGELO, TEXAS 31N

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS 5 (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
1310X	070869	27.2	1	3920	- 9.4	28.1	158.4	158.1	
			2	1500	- 3.7	27.9	157.4		
1311X	070969	32.2	1	370	- 8.5	26.7	150.6	150.6	A
			2	459	- 8.2				
1312X	071069	20.9	1	5060	-12.5	26.8	151.2	150.6	
			2	1025	0.0	26.2	147.8		
1314X	090469	21.1	1	6000	- 8.5	24.6	138.7	138.6	
			2	800	-----	24.4	137.6		B
1316X	090669	24.4	1	4800	- 8.2	26.3	148.3	149.8	
			2	1144	- 2.4	27.7	156.2		
1317X	091469	32.8	1	380	-----	24.7	139.3	156.0	C
			2	500	- 5.7	29.9	168.6		
1318X	091569	28.1	1	2900	-----	25.0	141.0	141.8	C
			2	1200	- 2.7	25.5	143.8		
1319X	091669	36.4	1	730	- 9.4	26.7	150.6		
			2	97	- 7.4	-----	-----		D
1320X	110169	20.8	1	4533	- 7.9	26.8	151.2	150.9	
			2	375		26.1	147.2		

A. BFD 1 AND 2 COMBINED FOR SPECIFIC ACTIVITY ANALYSIS

B. NO ANALYSIS OF δ C-13 FOR BED 2

C. NO ANALYSIS OF δ C-13 FOR BED 1

D. SPECIFIC ACTIVITY NOT COUNTED FOR BED 2

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS

SAN ANGELO, TEXAS 31N

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS 5 (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
1321X	110369	23.9	1 2	3500 580	- 9.2 + 0.1	26.6 27.1	150.0 152.8	150.4	
1322X	110569	32.3	1 2	525 340	- 9.2	25.1	141.6		
1323X	110669	26.2	1 2	2355 1112		23.8 24.7	134.2 139.3	135.8	
1326X	010770	21.4	1 2	2500 400	- 7.2 + 1.6	24.0 21.4	135.4 120.7	133.3	
1327X	010870	23.8	1 2	4200 185	- 6.6 + 0.2	25.8 -----	145.5 -----		A
1328X	011270	32.6	1 2	270 650	{ - 5.4	{ 23.4	132.0	132.0	B C
1329X	011670	27.2	1 2	2100 1100		26.6 30.2	150.0 170.3	156.9	
1331X	031270	21.5	1 2	3250 600		25.7 23.2	144.9 130.8	142.6	
1332X	031370	24.3	1 2	3350 730		27.4 24.0	154.5 135.4	151.1	

A. SPECIFIC ACTIVITY NOT COUNTED FOR BED 2

B. BED 1 AND 2 COMBINED FOR ANALYSIS

C. BEDS APPEARED TO HAVE BEEN RETURNED TO LABORATORY IN REVERSE

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS

SAN ANGELO, TEXAS 31N

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS 5 (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
1333X	031770	27.2	1	2500		25.1	141.6	148.5	
			2	1300		28.7	161.9		
1334X	032370	32.9	1	650		{23.9	{134.8	134.8	A
			2	440					
1337X	050770	32.6	1	730		21.8	123.0		
			2	95		----	-----		B
1346	090270	21.2	1	2140	- 4.6	24.0	135.4		
			2	175	- 0.6				
1347	090970	24.6	1	1940	-11.2	24.7	139.3	138.9	
			2	365	- 0.3	24.3	137.1		
1348	091170	32.6	1	476		{20.6	{116.2	116.2	A
			2	195	- 4.7				
1349	091870	32.7	1	350	-18.5	{22.7	{128.0	128.0	A
			2	280	- 9.6				
1350	091970	27.5	1	880		{25.2	{142.1	142.1	A
			2	400	- 7.8				
1351	092070	36.3	1	290	-13.5	{17.1	{96.4	96.4	A
			2	150	- 8.5				

A. BED 1 AND 2 COMBINED FOR SPECIFIC ACTIVITY ANALYSIS

B. SPECIFIC ACTIVITY NOT COUNTED FOR BED 2

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS
SAN ANGELO, TEXAS 31N

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS S (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
1352	110370	32.9	1	870	-11.3	23.2	130.8	129.9	
			2	250	- 4.8	22.5	126.9		
1353	110470	23.9	1	3600	- 8.4	25.2	142.1	141.6	
			2	1075	- 2.3	24.8	139.9		
1354	111070	21.2	1	4800	-11.5	22.7	128.0		
			2	500		-----	-----		A
1355	111770	27.3	1	2970	-10.2	23.7	133.7	130.8	
			2	1100	- 4.6	21.8	123.0		
1367	030371	21.5	1	3800	- 5.5	21.4	120.7	120.8	
			2	625	+ 0.9	21.5	121.3		
1368	031071	24.6	1	4300	- 6.8	22.0	124.1	123.3	
			2	940	+ 0.3	21.2	119.6		
1371	031771	27.7	1	3350	- 7.0	22.4	126.3	128.3	B
			2	1600	- 3.5	23.5	132.5		
1372	032271	27.9	1	2575	- 9.7	23.2	130.8	128.7	
			2	1500	- 6.2	22.2	125.2		
1376	052571	27.5	1	2975	- 8.4	20.7	116.7	116.7	
			2	1430	- 3.1	20.7	116.7		

A. LOST IN ANALYSIS

B. SAMPLER DAMAGED ON IMPACT, POSSIBLE CONTAMINATION

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS
SAN ANGELO, TEXAS 31N

FLIGHT NO.	DATE	ALT	BED NO.	VOL. CO-2	δ C-13 CONC.	EXCESS SPEC. ACT.	C-14 EXCESS	C-14 EXCESS WT.AVG.	R E M A R K S
		(KM)		(CC)	(PER MIL)	(DPM/GC)	⁵ (10 ATOMS/G AIR)		
1377	052671	32.4	1	925	-11.3	21.8	123.0	122.1	
			2	600	- 8.9	21.4	120.7		
1378	052871	24.6	1	3425	- 6.3	22.0	124.1	123.0	
			2	1250	- 1.7	21.3	120.1		
1381	060671	36.6	1	465		19.6	110.5	110.9	
			2	100		20.0	112.8		

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS

ALBROOK AFB, CANAL ZONE 9N

FLIGHT NO.	DATE	ALT	BED NO.	VOL. CO-2	δ C-13 CONC.	EXCESS SPEC. ACT.	C-14 EXCESS	C-14 EXCESS WT.AVG.	R E M A R K S
		(KM)		(CC)	(PER MIL)	(DPM/GC)	⁵ (10 ATOMS/G AIR)		
112	102770	31.5	1	1850	-11.0	12.8	-----		A
			2	410	- 3.4	18.7	105.5		
113	102870	27.1	1	2700	- 5.5	19.8	111.7	110.5	
			2	1400	- 3.4	19.2	108.3		
114	102970	20.7	1	4300	- 4.8	13.5	76.1	77.0	
			2	715	- 7.3	14.6	82.3		
115	103070	23.7	1	3400	- 8.0	21.4	120.7	120.3	
			2	700	- 2.7	21.0	118.4		
116	011371	25.5	1	2300	- 3.3	17.9	101.0	102.1	
			2	500	- 3.3	19.0	107.2		
117	011471	24.0	1	3750	- 7.3	19.2	108.3	108.1	
			2	850	- 2.8	19.0	107.2		
118	011671	21.1	1	4900	- 5.7	13.8	77.8	75.4	
			2	875	- 3.0	10.9	61.5		
119	012071	27.3	1	3100	- 9.0	19.7	111.1		
			2	950	- 4.4	-----	-----		B
120	012271	32.4	1	1350	-11.5	20.3	114.5	114.9	
			2	450	-16.5	20.6	116.2		

A. LEAK IN THE VACUUM SYSTEM, DATA SUSPECT.

B. LOST IN PURIFICATION.

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS
ALBROOK AFB, CANAL ZONE 9N

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS 5 (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
121	040371	21.4	1	5400	- 6.4	15.6	88.0	86.6	
			2	940	+ 0.1	13.9	78.4		
122	040471	24.4	1	3800	-14.2	17.2	97.0	98.0	
			2	1100	- 2.2	18.0	101.5		
123	040571	27.5	1	3000	- 8.2	20.4	115.1	115.1	
			2	1525	- 3.1	20.4	115.1		
124	060671	32.0	1	1010	-10.1	19.2	108.3	111.5	
			2	535	- 3.8	20.8	117.3		

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS
LONGREACH, AUSTRALIA 23S

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS 5 (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
511	012671	21.5	1 2	3200 195	- 5.1	25.1 -----	141.6 -----		A
512	012771	24.1	1 2	2840 600	- 3.4 - 6.0	22.3 19.2	125.8 108.3	122.8	
513	012971	27.3	1 2	1300 810	- 6.4 - 4.9	21.6 21.8	121.8 123.0	122.3	
514	020371	32.0	1 2	700 250	-10.2	19.4 -----	109.4 -----		A
522	042771	19.9	1 2	2710 250	-11.2 - 2.5	21.2 19.5	119.6 110.0	118.8	
523	042971	23.5	1 2						
524	043071	26.5	1 2	2500 380		22.3 22.0	125.8 124.1	125.6	
525	050371	31.1	1 2	1000 300	- 6.7	19.2 22.3	108.3 125.8	112.3	

A. NO ANALYSIS OF SPECIFIC ACTIVITY FOR BED 2.

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS
MILDURA, AUSTRALIA 345

FLIGHT NO.	DATE	ALT (KM)	BED NO.	VOL. CO-2 (CC)	δ C-13 CONC. (PER MIL)	EXCESS SPEC. ACT. (DPM/GC)	C-14 EXCESS 5 (10 ATOMS/G AIR)	C-14 EXCESS WT.AVG.	R E M A R K S
491	062370	27.3	1 2	2500 860	- 9.5 - 5.5	26.2 24.2	147.8 136.5	144.9	
492	070270	24.2	1 2	2400 460	- 8.9 - 6.9	25.2 19.6	142.1 110.5	137.0	
493	070770	21.2	1 2	2650 250	- 7.5 -10.2	26.3 25.9	148.3 146.1	148.1	
495	091670	----	1 2	3500		12.7	71.6		A
496	091770	24.0	1 2	2370 135	- 6.0 - 0.1	24.0	135.4		
497	091870	20.8	1 2	2300 570		23.1 21.3	130.3 120.1	128.3	
498	093070	27.4	1 2	1133 220	- 3.7 - 3.2	21.2 21.5	119.6 121.3	119.9	
499	100270	----	1 2	1960	- 4.6	11.9	67.1		B
507	121270	21.0	1 2	3000 335	- 3.4 - 3.5	21.2 20.7	119.6 116.7	119.3	

A. BALLOON FAILURE. SAMPLED FROM 32KM TO SURFACE.

B. BALLOON FAILURE. SAMPLED FROM 36KM TO 6KM.

TABLE 1 STRATOSPHERIC CARBON-14 MEASUREMENTS
MILDURA, AUSTRALIA 34S

FLIGHT NO.	DATE	ALT	BED NO.	VOL. CO-2	δ C-13 CONC.	EXCESS SPEC. ACT.	C-14 EXCESS	C-14 EXCESS WT.AVG.	R E M A R K S
		(KM)		(CC)	(PER MIL)	(DPM/GC)	⁵ (10 ATOMS/G AIR)		
508	121370	24.2	1	1700	- 1.9	22.7	128.0	130.0	
			2	860	- 8.1	23.9	134.8		
509	121570	27.1	1	1920	- 8.3	24.2	136.5	133.6	
			2	260		19.9	112.2		
515	031171	23.4	1	3070	- 6.6	23.4	132.0	132.6	
			2	700	- 3.8	24.0	135.4		
516	031671	26.8	1	1900	- 7.6	23.6	133.1	131.4	
			2	530	- 3.6	22.2	125.2		
518	032371	20.9	1	3150	-10.0	23.2	130.8	130.0	
			2	265	+ 1.7	21.4	120.7		
519	032471	32.8	1	915	-12.3	22.6	127.5	128.5	
			2	260	- 4.9	23.4	132.0		
521	040571	36.5	1	550	-12.3	20.3	114.5	114.6	
			2	165	- 3.2	20.4	115.1		

Table 2. Argonne National Laboratory
 δ C-13 (0/00) Determinations.

<u>Collector</u>	<u>Average</u>	<u>Standard Deviation</u>	<u>No. Cases</u>
1. Molecular Sieve			
Bed-1 (Primary)	-8.5	3.1	81
Bed-2 (Breakthrough)	-4.5	4.0	77
Wt. Avg. (Bed 1 and 2)	-7.7	2.7	72
2. Whole Air	-8.1	1.0	99

**Table 3. Stratospheric Burden of Excess
C-14 (10^{26} atoms) from 21 to 37 km.**

	Sept. - Dec. 1970	Jan. - June 1971
N.H.	12.8	12.1
S.H.	12.4	12.6
Total	25.2	24.7
Estimated*		
Total	16	15

***Assumes an effective half-residence time of 60 months between June 1964 to July 1966.**

Fig.1: Schematic Diagram Of The Molecular Sieve Sampler (Ashenfelter et al, 1972)

Fig 2. Vertical Distribution of δC^{13} (per mil)

Fig 3. Vertical Distribution of δC^{13} (per mil)

Fig 5. Excess C-14 (10^5 atoms/grams of air)
January - June, 1971

by B. G. Bennett (HASL)

ABSTRACT

From the long term HASL programs of Sr-90 deposition measurement and analysis of Sr-90 in milk and total diet in New York City, methods have been developed for making reliable estimates of Sr-90 levels in milk and total diet. The deposition-milk relationship includes a lag term to account for use of stored feed and exponential removal of the contribution from deposition of preceding years. Exponential removal, to account for decay and reduced uptake availability of Sr-90 in soil, amounts to 13% per year in New York. The diet-milk ratio, commonly used to estimate dietary intake of Sr-90 from Sr-90 intake in milk, varies considerably about 1.4 for New York and would be expected to vary similarly in other areas through periods of changing deposition. Improvements in dietary Sr-90 intake estimates are achieved by using quarterly milk measurements and estimating Sr-90 intake in non-dairy foods from the Sr-90 intake in milk in the preceding quarter.

The Health and Safety Laboratory began a program to measure global Sr-90 fallout in 1954, shortly after large scale atmospheric testing began to introduce significant amounts of Sr-90 into the environment. The continuous record of Sr-90 deposition measurements in New York City is shown in Figure 1. Monthly analyses of Sr-90 in milk from New York City also began in 1954 (Figure 2). Total dietary Sr-90 intake in New York City, estimated from quarterly analyses

Fig. 1. Monthly Sr-90 deposition in New York City.

Fig. 2. Monthly Sr-90 concentration in milk in New York City.

Fig. 3. Quarterly dietary Sr-90 intake in New York.

of 19 representative food items, has been reported since 1960 (Figure 3).

From these relatively long-term monitoring programs, it is possible to make fairly good correlations between Sr-90 deposition and the levels in milk and total diet. Estimation of the transfer of Sr-90 fallout to milk and diet under varying conditions of deposition can now be made rather reliably. This report updates the experience gained from these studies.

Correlation of Sr-90 Deposition with Levels in Milk

The Sr-90 levels in milk have traditionally been described by a model which relates the yearly average level in milk to the fallout rate in the current year and the cumulative deposit in the soil. The model, sometimes known as the UN model because of its use by the United Nations Scientific Committee on the Effects of Atomic Radiation, was devised by Tajima.⁽¹⁾

As the model developed, a lag factor to account for use of stored feed was added, and most recently, exponential removal has been included in the cumulative deposit term to account for losses due to downward movement in soil and other physical and chemical changes which reduce the availability

of Sr-90 for uptake by plants. O'Brien has proposed such an updated formula for use by the UN committee. ⁽²⁾

The model which best describes the New York data is given by the following formula:

$$M_n = p_1 F_n + p_2 F_{n-1} + p_3 \sum_{m=1}^{\infty} F_{n-m} e^{-m\lambda}$$

$$p_1 = .69 \qquad p_2 = .21 \qquad p_3 = .20 \qquad \lambda = .14$$

In this model p_1 is the proportionality constant which includes the rate and deposit effects of the current year's deposition, p_2 is the lag factor for the previous year's deposition, and $p_3 e^{-m\lambda}$ is the deposit factor with exponential removal of the deposition in each of the preceding years. M_n (pCi/gCa) is the average Sr-90/Ca ratio in milk in the year n , and F_n (mCi/km²) is the Sr-90 fallout deposition during the year n . Figure 4 shows the least squares fit to the average yearly Sr-90 concentrations in milk. Estimates from the model are usually within 10% of the average observed values.

The model above is just slightly modified from the formulation proposed by Bartlett and Russell ⁽³⁾ and applied by them to UK data. The Bartlett-Russell formulation splits the preceding year's deposition and lags only the last half of

Figure 4. Average yearly Sr-90 concentration in milk in New York City.

the year. The lag effect is less important in New York ($p_2 = .21$) than in the UK ($p_2 = 1.41$, equivalent to .70 for lag of the entire previous year's deposition), which indicates less use of stored feed in the New York area than in the UK. The other parameters are in close agreement ($p_1 = .69$ and .70, $p_3 = .20$ and .20, $\lambda = .14$ and .18 in New York and in the UK, respectively).

A model applied to Norwegian data by Hvinden and Lillegraven⁽⁴⁾ and another model applied to Italian data by Van der Stricht, Gaglione, and de Bartoli⁽⁵⁾ include exponential removal but no lag term. The mean residence times of Sr-90 in soil inferred from the exponential removal terms vary in these analyses from 1.9 years in Norway to 5.6 years in the UK and 6.7 years in Italy. This may be compared to 7.1 years determined for New York, corresponding to 11% per year removal in addition to the 2.4% radioactive decay.

The Diet-Milk Ratio

A common practice to estimate total dietary intake of Sr-90, short of sampling many more food items, has been to multiply the Sr-90/Ca ratio measured for milk by a constant factor, usually 1.4, though 1.0 has been found more appropriate for the UK and values from 2 to 4 are needed in the USSR and Japan due to differences in the diets.

$$\text{Diet (pCi/gCa)} = 1.4 \times \text{Milk (pCi/gCa)}$$

The factor 1.4, as an average value for recent years, also applies to New York. A careful look at the variations in this ratio, however, show changes in time which can quite reasonably be expected.

For typical western diets, milk and other dairy products contribute ~60% of the dietary calcium intake. The diet-milk ratio can thus be written.

$$\left(\frac{\text{Sr-90}}{\text{Ca}}\right)_{\text{diet}} \bigg/ \left(\frac{\text{Sr-90}}{\text{Ca}}\right)_{\text{milk}} = .6 \times (\text{Sr-90})_{\text{diet}} / (\text{Sr-90})_{\text{milk}}$$

Milk is one of the first food items contaminated following a period of deposition. If milk is the only food contaminated, the diet-milk ratio would be .6. As the contamination begins to appear in other foods, the diet-milk ratio increases. When milk contributes just over 40% of the dietary Sr-90 intake, the diet-milk ratio is 1.4. The variations in the contribution of milk to the total diet Sr-90 intake reflect the variations in amounts of Sr-90 deposition.

Figure 5 shows the changes in the diet-milk ratio for New York from 1961 to 1970. During periods of heavy fallout deposition, 1962-64, the milk contribution was greater than 40%⁽⁶⁾, and the ratio was less than 1.4. As the levels in grain and vegetables become more important relative to milk, the contribution from milk fell below 40%, and the diet-milk ratio increased above 1.4. In recent years the ratio has been stabilizing around 1.4.

Fig. 5. Diet-milk ratio in New York City.

The use of a constant diet-milk ratio, such as 1.4, leads to yearly dietary intake estimates which are correct to within about $\pm 50\%$, but much better estimates of total diet Sr-90 intake can be obtained by adjusting the diet-milk ratio according to the level of deposition. Adjustments similar to those shown in Figure 5 should be helpful.

Estimation of Total Diet Sr-90 Intake from Milk Levels

It would seem that even better use could be made of milk levels to estimate the total diet Sr-90 intake than the use

of the diet-milk ratio applied to yearly averages. One might, first of all, make use of the quarterly values of Sr-90 in milk, which should usually be available. In obtaining representative average yearly milk levels, measurements should have been made on at least a quarterly basis. One then requires a relation between quarterly intake of Sr-90 in milk and quarterly intake of Sr-90 in other, non-dairy, foods. This relation is not so direct, however, since there is a lag in contamination of other foods compared to milk following Sr-90 deposition. The quarterly results for milk and other foods from New York shown in Figure 3 show quite clearly that the lag is about 1 quarter (3 months). Thus, the quarterly intake of Sr-90 in milk should best be used as an indicator of the Sr-90 intake in other foods in the succeeding quarter.

$$D_i = M_i + f \times M_{i-1}$$

D_i = total diet Sr-90 intake in i^{th} quarter (pCi)

M_i = Sr-90 intake in milk in i^{th} quarter (pCi)

f = the factor relating quarterly intake of Sr-90 in milk to the quarterly intake of Sr-90 in other foods in the following quarter.

It is seen from Figure 3 that the contribution from other foods to the total Sr-90 intake is greater than the milk con-

tribution, except during the periods of heavy deposition in 1962-63 and in the spring of 1964. Thus even this method of estimation of Sr-90 intake must be adjusted when the deposition is particularly heavy.

Figure 6 shows, for the New York data, how the factor f varied during the period 1960-70 in relation to the total Sr-90 deposition during the quarter. For low quarterly deposition f varies from 0.8 to 2, the average value for quarterly deposition less than 3 mCi/km^2 being 1.4. Above 3 mCi/km^2 the factor decreases. For prediction purposes, a value of $f = 1.0$ for deposition from 3 to 5 mCi/km^2 and $f = 0.8$ for deposition $\geq 5 \text{ mCi/km}^2$ would appear suitable.

Fig. 6. Variation, relative to quarterly Sr-90 deposition, of the ratio of quarterly intake of Sr-90 in other foods to that of milk in the previous quarter.

The quarterly deposition, referred to Figure 6, has been shifted one month relative to the quarterly milk intake, since in comparing the deposition and milk records of Figures 1 and 2, it is noted that peak deposition usually precedes peak levels in milk by about 1 month. Thus, for example, in using the first quarter milk intake (Jan., Feb., Mar.) to estimate second quarter intake of other foods (Apr., May, June), the choice of f depends on the Sr-90 deposition in the first quarter shifted one month (Dec., Jan., Feb.). The one month shift in quarterly deposition is not too important, however, and it usually makes very little difference in the choice of f.

The estimation method may be summarized by the following relations:

Deposition

Dietary Intake

$$< 3 \frac{\text{mCi}}{\text{km}^2 \cdot \text{Qtr}}$$

$$D_i = M_i + 1.4 M_{i-1}$$

$$3-5 \frac{\text{mCi}}{\text{km}^2 \cdot \text{Qtr}}$$

$$D_i = M_i + 1.0 M_{i-1}$$

$$\geq 5 \frac{\text{mCi}}{\text{km}^2 \cdot \text{Qtr}}$$

$$D_i = M_i + .8 M_{i-1}$$

D_i = Sr-90 intake in total diet in quarter i (pCi)

M_i = Sr-90 intake in milk in quarter i (pCi)

Using these relationships we may show in Figure 7 how the observed diet levels from 1960-70 are reproduced. The estimates of total diet intake obtained from the milk levels measured from 1954 to 1960 are also included in this figure. The dashed lines show the quarters in which the deposition was $> 3 \text{ mCi/km}^2$, thus requiring adjustment in the factor f . The solid lines above the dashed lines show the overestimate obtained if the factor is not adjusted. Even without adjustment, the yearly intake estimated on this quarterly basis is within 25% of the observed values. The adjusted quarterly total diet Sr-90 intake estimates are quite close to the observed results for the entire sampling period.

In terms of the Sr-90/Ca ratios in diet and milk, the estimation formula for deposition $< 3 \text{ mCi/km}^2 \cdot \text{Qtr}$ becomes (based on 386 gCa/yr in diet and 222.6 gCa/yr in milk):

$$\text{Quarterly} \quad D_i = .6 M_i + .8 M_{i-1}$$

$$\text{Yearly} \quad D_n = \frac{1}{4} \{ .8 M_0 + 1.4 (M_1 + M_2 + M_3) + .6 M_4 \}$$

When M_0 , the Sr-90/Ca ratio in milk in the 4th quarter of the year $n-1$, is equal to M_4 , the ratio in the 4th quarter of the year n , that is when milk levels are unchanging,

Fig. 7. Quarterly Sr-90 intake in total diet in New York City.

$$D_n = 1.4 M_n$$

where D_n and M_n are the average Sr-90/Ca ratios in the diet and in milk in the year n .

This, of course, brings us back to the use of the average diet-milk ratio, but only when the deposition is low. In periods of heavy Sr-90 deposition, the dietary Sr-90 intake estimates should be adjusted, as discussed above, and, if Sr-90 levels in milk are changing, quarterly estimation of dietary Sr-90 intake is recommended.

Conclusion

The long-term sampling programs conducted by this laboratory have permitted quite good correlation of Sr-90 fallout deposition with the Sr-90 levels in milk and have indicated a relationship between Sr-90 intake in milk and in total diet which should allow projections or estimates of Sr-90 intake, where sampling is not as complete, to be made with confidence.

The estimation techniques have been derived from the New York data, but the methods should be generally applicable. The parameters for other areas might be expected to vary slightly because of different soil conditions and pasturing practices and also differences in diets. In instances where

the data is less localized, such as country-wide survey measurements, some loss of detail might be expected.

Brief investigation of the diet and milk data variously reported from other countries has also shown variations of the diet-milk ratio with time. In most areas the ratio decreases, as would be expected, during the 1962-64 heavy fallout period. Those countries that no longer analyze total diet or that have traditionally analyzed only milk may wish to consider some adjustment of the diet-milk ratio to account for its non-constant nature. When adjustment of the diet-milk ratio is undertaken, or even when it is not, better estimates of dietary Sr-90 intake are obtained when quarterly results from milk programs are used in the manner described above.

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PART II

HASL FALLOUT PROGRAM DATA

1. Fallout Deposition

1.1 Monthly Precipitation

1.11 Sr^{90} and Sr^{89} in Monthly Deposition at World Land Sites

Precipitation and dry fallout are collected over monthly periods at stations in the United States and overseas. The samples are analyzed for Sr^{90} , and Sr^{89} whenever possible. A description of the sampling network and available data for each site are given in the Appendix, Section A.

1.12 Other Isotopes at Selected Sites

At a number of stations in the United States, monthly deposition collections were analyzed for radiostrontium and other nuclides of interest to the Atomic Energy Commission. Multinuclide analyses were discontinued as of July 1967 and the complete data reported in HASL-193, p. II-4 thru II-25.

Plutonium analyses of monthly deposition were carried out at New York City, Melbourne, Seattle, Honolulu, Salisbury, Durban, and Rio de Janeiro. This program has been terminated and the complete data are given in HASL-237 on pages II-4 thru II-13.

1.2 Sr^{90} and Sr^{89} Deposition at Atlantic Ocean Weather Stations

Measurements of radiostrontium in precipitation and dry fallout collections at four U. S. Coast Guard Stations in the North Atlantic Ocean are carried out for comparison with land stations in the same latitude band. A description of the stations and available data are given in the Appendix, Section B.

2. Surface Air Sampling Program

The Health and Safety Laboratory has been collecting surface air particulate samples at stations in the Western Hemisphere since January 1963. The filters are analyzed for a number of fission and activation product radionuclides as well as stable lead. A description of the program and available data are given in the Appendix, Section C.

3. Project Airstream

The Health & Safety Laboratory measures radioactivity in the lower stratosphere employing the RB-57F aircraft as a sampling platform. The aircraft are flown by the 58th Weather Reconnaissance Squadron under the direction of the 9th Weather Wing of the Air Weather Service. The missions are scheduled quarterly and the coverage extends from 75°N to 51°S latitude in the Western Hemisphere. Air filter samples are collected from 12 to 19 km altitude and analyzed for a number of radionuclides. A more complete description of the program and available data are given on pages II-8 to II-36 of HASL-245.

4. High Altitude Balloon Sampling Program

Balloon borne filtering devices are used to collect nuclear debris at altitudes from 21-32 km. Balloon launchings are conducted annually at Fairbanks, Alaska, 65°N; Sioux City, Iowa 42°N; Alamogordo, N.M., 33°N; and Panama C.Z., 9°N and quarterly at Mildura, Australia, 34°S. Filters are analyzed for a number of radionuclides. A more complete description of the program and available data are given on pages II-37 to II-93 of HASL-245.

5. Radiostrontium in Milk and Tap Water

Sr⁹⁰ levels in both powdered and fresh milk distributed in New York City and tap water sampled at the Health & Safety Laboratory, have been measured on a monthly basis since 1954. These data are summarized in tabular and graphical form in the Appendix, Section D.

6. Strontium⁹⁰ in Diets

Quarterly estimates of the annual dietary intake of Sr⁹⁰ of New York City and San Francisco residents have been made based on analyses of foods purchased at these cities every three months since 1960. Sampling in Chicago was discontinued in 1967. The program is described and available data reported on p. II-4 to II-6 of this report.

7. UNSCEAR - WHO Bone Program

Available Sr⁹⁰ data for human bone samples collected in 1969, 1970, and 1971 from countries in Latin America, Africa, and Asia are presented on pages II-4 to II-7 of HASL-245.

8. Sr⁹⁰ in Human Bone - 1970 Results

Specimens of human vertebrae from New York City and San Francisco have been made available for Sr⁹⁰ analysis since 1961. The results for 1970 are evaluated beginning on page I-99 of HASL-243.

6. HASL Diet Studies: Second Quarter 1971

Results of the measurements of the Sr^{90} content of foods purchased in New York City and San Francisco in the second quarter of 1971 are given in the following table. Estimates of the daily intake of Sr^{90} based on these measurements and on the revised consumption statistics given in the most recently available U. S. Department of Agriculture Report (1), are also listed.

The estimates of daily Sr^{90} intake are a continuation of the HASL Tri-City diet studies which were started in March of 1960. Results of the earlier measurements along with those made during 1970 are shown graphically in the figure on page II-6. A complete description of the sampling methods and philosophy of the HASL diet studies was given in HASL-147(2). A discussion of the reasons for changes in the Tri-City diet studies made in 1968 was given in HASL-200(3).

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- (1) Food Consumption of Households in the United States - Spring 1965
A Preliminary Report, U.S.D.A. ARS 62-16, August 1967
- (2) Rivera, J. and Harley, J. H.
HASL Contributions to the Study of Fallout in Food Chains
USAEC Report HASL-147, July (1964)
- (3) USAEC Report HASL-200, October (1968), p. II-6

STRONTIUM⁹⁰ IN NEW YORK CITY AND SAN FRANCISCO DIETS
- Second Quarter 1971 -

Diet Category	kg/yr	gCa/yr	% of yearly intake of Ca	NEW YORK CITY - MAY			SAN FRANCISCO - JUNE		
				pCi Sr ⁹⁰ kg	pCi Sr ⁹⁰ yr	% of yearly intake of Sr ⁹⁰	pCi Sr ⁹⁰ kg	pCi Sr ⁹⁰ yr	% of yearly intake of Sr ⁹⁰
DAIRY PRODUCTS	200	216.0	58	9.3	1860	37	2.9	583	36
FRESH VEGETABLES	48	18.7		19.0	914		2.0	95	
CANNED VEGETABLES	22	4.4		2.3	50		3.1	68	
ROOT VEGETABLES	10	3.8		6.3	63		3.3	33	
POTATOES	38	3.8		6.8	259		0.6	21	
DRY BEANS	3	2.1	9	8.4	25	26	17.2	52	16
FRESH FRUIT	59	9.4		14.2	840		2.6	151	
CANNED FRUIT	11	0.6		1.6	18		1.7	19	
FRUIT JUICES	28	2.5	3	3.5	99	19	2.8	79	15
BAKERY PRODUCTS	44	53.7		8.5	372		3.8	166	
FLOUR	34	6.5		7.7	263		4.9	166	
WHOLE GRAIN PRODUCTS	11	10.3		13.2	145		6.5	71	
MACARONI	3	0.6		4.5	14		3.6	11	
RICE	3	1.1		1.9	6		2.4	7	
			20			16			26
MEAT	79	12.6		0.6	48		0.7	52	
POULTRY	20	6.0		0.9	19		1.1	21	
EGGS	15	8.7		2.2	33		2.3	34	
FRESH FISH	8	7.6		1.2	10		0.3	2	
SHELLFISH	1	1.6	10	1.1	1	2	0.4	<1	7
YEARLY INTAKE		370			5039			1632	
DAILY INTAKE = pCi per gCa					13.6			4.4

PART III

DATA FROM SOURCES OTHER THAN HASL

Numerous fallout studies are conducted by other organizations in the United States and abroad. Some of these are sent to the editors for dissemination in these HASL Quarterly Reports. Submitted data are reproduced essentially as received and no interpretation by HASL is attempted.

	<u>Page</u>
1. EURATOM Joint Nuclear Research Centre Ispra Establishment, Protection Service Site Survey and Meteorology Section, Quarterly Report	III-2
2. Radiological Physics Division, Argonne National Laboratory Cesium-137 in Various Chicago Foods (Collection Month, October 1971) by S. S. Brar and D. M. Nelson	III-7
3. Global Atmospheric Radioactivity: January 1970	III-11

ISPRA ESTABLISHMENT

Protection Service

Site Survey and Meteorology Section

QUARTERLY REPORT

EUR/C-15/394/70e.

The Euratom Ispra Establishment is located in Northern Italy 58 Km NW from Milan and 14 Km W from Varese.

The activity levels shown in this report represent world wide fallout, and do not reflect any contamination from the site.

SAMPLE COLLECTION

a. Air

Air is drawn by pumps through paper filters at the rate of, at least, 500 m³/day, measured by gas meter.

The single daily filters are measured for gross beta radioactivity and then pooled to give monthly samples, for gamma spectrometry and radiochemical analyses.

b. Wet and dry deposition

These samples are collected monthly by means of four 1 m² stainless steel funnels, with the bottom always covered with deionized water. Water in excess of a pre-fixed level drains through a filter and a double (cationic and anionic) ion-exchange column. The eluate of the resins and the filters are submitted to the analyses. This procedure is described in detail in the paper by M.C. de Bortoli and P. Gaglione : "Sampling of radioactive deposition through ion-exchange resins", *Giornale di Fisica Sanitaria*, 13, 219-224 (1969).

c. Milk

Milk is collected twice a week in four small local dairies to give 8 liters /month. About six liters are dried and submitted to gamma spectrometry and two liters ashed for radiochemical determination of strontium-90.

CHEMICAL PROCEDURES AND COUNTING TECHNIQUES

- a. Strontium-90 is separated by the fuming nitric acid precipitation and then purified through hydroxides and chromates precipitations. The activity of the final strontium carbonate and yttrium oxalate precipitates is measured in low level anticoincidence beta counters.
- b. Cesium-137 is measured by direct gamma spectrometry on the unprocessed or dried samples and, whenever it is necessary, by gamma spectrometry after chemical separation. This is performed by filtration of the solution, obtained dissolving the sample, through a thin AMP (ammonium molybdophosphate) layer, by which cesium is retained. Details of this procedure may be found in the paper by L. Van der Stricht issued on "Radiochimica Acta" 3, 193-199 (1964).
- c. Gamma emitting nuclides are measured by direct gamma spectrometry, using, also the spectrum stripping technique.
- d. Plutonium isotopes are separated by anion exchange and electro-deposition ; details of the procedure may be found in the paper by M.C. de Bortoli : "Radiochemical determination of plutonium in soil and other environmental samples", Anal. Chem. 39, 375 (March 1967).
The activity is measured in a Frish grid ionisation chamber connected to a multichannel analyser.

EXTRAPOLATION OF THE DATA

Except when otherwise stated, the data presented in this report are extrapolated to the last day of the collecting period.

AIR RADIOACTIVITY

1971

SITE : I S P R A

LAT. 45° 49' N

LONG. 8° 37' E

ALT. 250 m

Month	Gross beta pCi/m ³	⁹⁰ Sr 10 ⁻³ pCi/m ³	⁸⁹ Sr 10 ⁻³ pCi/m ³	¹³⁷ Cs 10 ⁻³ pCi/m ³	²³⁹ Pu 10 ⁻⁵ pCi/m ³	²³⁸ Pu 10 ⁻⁵ pCi/m ³
June	0.68	5.5	27	8.4	12	0.66
July	0.70	6.6	19	9.0	12	0.62
August	0.41	4.0	4.5	6.1	6.1	0.34
September	0.21	1.7	1.5	2.6	-	-

- Data not yet available

FALLOUT DEPOSITION

1971

SITE : I S P R A

LAT. 45° 49' N

LONG. 8° 37' E

ALT. 250 m

Month	Gross beta (1)		⁹⁰ Sr	⁸⁹ Sr	¹³⁷ Cs	²³⁹ Pu	²³⁸ Pu	Precipitation mm
	mCi/Km ²	Date (2)	mCi/Km ²	mCi/Km ²	mCi/Km ²	μCi/Km ²	μCi/Km ²	
January	2.9	15-2	0.070	0.23	0.096	0.51	0.046	121.6
February	1.4	10-3	0.023	0.091	0.040	0.29	0.015	74.2
March	6.2	20-4	0.063	0.57	0.14	0.70	0.064	154.8
April	23.8	13-5	0.31	2.63	0.55	3.1	0.34	180.8
May	39.2	22-6	0.67	5.68	0.97	10	0.72	316.8
June	40.2	8-7	0.65	3.03	1.12	14	0.77	233.0
July	12.2	6-8	0.21	0.60	0.31	3.3	0.17	59.0
August	13	3-9	0.25	0.50	0.35	3.9	0.21	92.2
September	2.0	5-10	0.03	0.03	0.057	-	-	13.4

(1) Potassium-40 equivalent (40 mg/cm²).

(2) Day and month of the gross beta measurement.

- Data not yet available.

Cesium-137 in Various Chicago Foods*
(Collection Month October, 1971)

S. S. Brar and D. M. Nelson
Radiological Physics Division
Argonne National Laboratory
Argonne, Illinois 60439

Since April, 1961, the ^{137}Cs and potassium content of the Chicago portion of Tri-City Diet Sampling Program has been determined¹⁻⁴ in bulk food samples by gamma ray spectrometry using a 4" x 4" NaI (TI) crystal. The individual food components were counted for a minimum of 100 minutes, and from these measurements composite daily and yearly food intakes were obtained. Cesium-137 activity in food, now, is an order of magnitude lower than it was a few years ago; consequently, a new procedure for these measurements has been adopted in order to improve accuracy. The same variety of foods (all fresh vegetables, all fresh fruits, etc.) are composited before measurement, and the samples are counted a minimum of 400 minutes. The results for the October, 1971 quarter are tabulated in Tables I and II.

*Work performed under the auspices of the U. S. Atomic Energy Commission.

Table 1

Cesium-137 in Chicago Diets

(Adults)

October, 1971

	kg/yr	Potassium g/kg	^{137}Cs pCi/kg	Potassium g/yr	^{137}Cs pCi/yr
White Bread	37	1.4	22	52	814
Whole Wheat Bread	11	2.6	37	29	407
Eggs	16	1.4	0	22	0
Fresh Vegetables	43	3.9	0	168	0
Root Vegetables	17	2.9	0	49	0
Milk	221	1.5	9	332	1989
Poultry	17	2.5	10	43	170
Fresh Fish*	8	3.0	32	24	256
Flour	43	1.1	28	47	1204
Macaroni	3	2.0	30	6	90
Meat	73	2.9	19	212	1387
Dried Beans	3	11.8	20	35	60
Fresh Fruit	68	1.9	6	129	408
Potatoes	45	4.2	0	189	0
Canned Fruits	26	0.9	9	23	234
Canned Fruit Juices	19	1.6	12	30	228
Canned Vegetables	20	0.7	8	<u>14</u>	<u>160</u>
Total/yr				1404	7407
Total/day				3.8	20

*It is assumed in arriving at the average that nine times more ocean fish is consumed than fresh water fish.

Table II

Cesium-137 in Chicago Diets
(Infants)

October, 1971

	kg/yr	Potassium g/kg	¹³⁷ Cs pCi/kg	Potassium g/yr	¹³⁷ Cs pCi/yr
Evaporated Milk	137	3.0	31	411	4247
Formula Milk	37	1.9	15	70	555
Cereals	8	2.7	23	22	184
Fruits	23	0.8	10	18	230
Meats	17	2.3	42	39	714
Vegetables	23	2.1	0	<u>48</u>	<u>0</u>
Total/yr				608	5930
Total/day				1.7	16

References

1. S. S. Brar, et al., USAEC Report No. HASL-146, Cs-137 in Various Chicago Diets, pp. 225-232, July 1, 1964.
2. J. Rivera and J. J. Kelly, USAEC Report No. HASL-144, Cs-137 in Tri-City Diets, pp. 228, April 1, 1964.
3. J. Rivera and J. H. Harley, USAEC Report No. HASL-147, Contributions to the Study of Fallout in Food Chains, pp. 31-35, July, 1964.
4. S. S. Brar and D. M. Nelson, USAEC Report No. HASL-245, Cs-137 in Various Chicago Foods, pp. III-25 to III-28, October 1, 1971.

GLOBAL ATMOSPHERIC RADIOACTIVITY

JANUARY 1970

Introduction

In January 1970, USAF aircraft were deployed to make a series of vertical profiles of particulate radioactivity in the atmosphere. Aircraft sampling was normally conducted in the vicinity of four latitudes: 70°N, 35°N, 10°N, and 40°S. Altitudes sampled varied from approximately 12.2 km (40,000 feet) to about 20.1 km (66,000 feet). Radiochemical analysis was by government laboratories.

These data were collected as part of a cooperative effort by the U.S. Department of Defense, Atomic Energy Commission and National Oceanic and Atmospheric Administration, and are a continuation of similar sampling programs conducted in the past (1-8).

Data

A summary of the data is given in the tables.

Date	Date (GCT) on which the filter was extracted from the aircraft, immediately after landing.
Altitude	Pressure altitude (in kilometers) at which sampling was performed.
Tropopause	Height of tropopause (in kilometers) as reported by a nearby radiosonde station.
Isotopic data	All data are reported in units of pico-curies per 100 standard cubic meters of air. All radioactivity data are corrected

for decay to day of extraction of the filter. The following constants were used:

Isotope	Half-life Days	Decay Constant Days ⁻¹
Be-7	53.0	0.0131
Mn-54	309	0.00224
Fe-55	1073	0.000646
Sr-89	53.0	0.0131
Sr-90	10104	0.0000686
Zr-95	63.6	0.0109
Cs-137	9708	0.0000714
Ba-140	12.8	0.0542
Ce-141	32.5	0.0213
Ce-144	282	0.00246
Pb-210	8030	0.0000863
Pu-238	31500	2.2×10^{-5}
Pu-239	8800000	7.8×10^{-8}

Unless otherwise indicated, the precision of the radiochemical analysis is within 3%. Standard errors in the analysis greater than 3% are indicated by a lower case letter following the value in accordance with the following code:

- a 3-10%
- b 11-20%
- c 21-40%
- d 41-100%
- e > 100%

NR indicates the isotope was not detected in resolvable amounts.

Acknowledgments

This program would not have been possible without the cooperation of the Government of Argentina.

References

1. Fallout Program, Quarterly Summary Report, Health and Safety Laboratory New York Operations Office, U.S.A.E.C., HASL-115, October 1, 1961, pp. 177-183.
2. _____, HASL-117, December 30, 1961, pp. 225-229.
3. _____, HASL-142, January 1, 1964, pp. 272-276.
4. _____, HASL-165, January 1, 1966, pp. 301-311.
5. _____, HASL-172, July 1, 1966, pp. 164-171.
6. _____, HASL-182, July 1, 1967, pp. III30-III47.
7. _____, HASL-214, October 1, 1969, pp. III12-III43.
8. _____, HASL-227, July 1, 1970, pp. III12-III21.

Atmospheric Radioactivity Data, January 1970

Latitude	70-75°N	70-75°N	70-75°N	67-70	70-75°N
Longitude	144°W	144°W	144°W	144°W	143°W
Date	13	20	13	13	12
Altitude (km)	12.2	12.2	15.2	15.2	18.3
Tropopause Ht. (km)	8.5-9.1	8.2-9.4	9.5-10.6	8.5-9.1	9.5-10.6
Isotopes					
Be-7	5850	2890	4660	19000	14700
Mn-54	8.9a	10a	31	63	13a
Fe-55	32b	--	--	190a	--
Sr-89	1160	1770	8380	11300	5240
Sr-90	56	100a	235b	274	183a
Zr-95	2120	3320	15700	20500	9950
Cs-137	97	82	278	509	272
Ba-140	177b	161	999	1640	688
Ce-141	--	1940	9210	--	6370
Ce-144	1370	2210	7930	12900	5170
Pb-210	0.93	< 0.3	< 0.7	0.61a	< 0.8
Pu-238	0.17a	0.23a	0.35a	0.39a	0.30a
Pu-239	0.69	0.99	2.6	3.5	1.7

Atmospheric Radioactivity Data, January 1970

Latitude	66-70°N	70-75°N	70-75	30-35°N	30-35°N
Longitude	145°W	144°W	144°W	103-107°W	103-106°W
Date	19	5	12	6	20
Altitude (km)	18.3	18.9	18.9	12.2	12.2
Tropopause Ht. (km)	9.1-9.8	6.8-11.0	9.5-10.6	10.7	12.2
Isotopes					
Be-7	4340	7530	25400	6460	818
Mn-54	21	13	21	3.8a	< 0.8
Fe-55	75a	33a	--	--	< 6
Sr-89	2930	2070	2780	493	25
Sr-90	114	67	92a	42a	2.6
Zr-95	5260	3590	5170	914	68
Cs-137	179	89	132	67	4.1
Ba-140	254a	331a	362	74a	NR
Ce-141	--	--	3310	547	--
Ce-144	3660	2030	2780	785	39
Pb-210	0.29b	0.17c	< 1	< 0.2	0.24b
Pu-238	0.35a	0.26a	0.30a	0.12a	0.02C
Pu-239	1.1	0.72	0.97	0.44	0.04

Atmospheric Radioactivity Data, January 1970

Latitude	30-35°N	30-35°N	30-35°N	30-35°N	30-35°N
Longitude	103-107°W	102-106°W	103-106°W	103-106°W	103-107°W
Date	5	12	5	19	12
Altitude (km)	15.2	15.2	18.3	18.3	19.2
Tropopause Ht. (km)	9.4-12.8	10.1-10.7	9.4-12.8	11.0-11.3	10.1-10.7
Isotopes					
Be-7	6670	2600	7770	9080	8960
Mn-54	11	1.9a	19a	36	31
Fe-55	--	6.5b	--	106a	--
Sr-89	2670	197	5360	5770	8500
Sr-90	97	11	181	197	302a
Zr-95	5190	352	9700	10300	16000
Cs-137	157	19	290	287	393a
Ba-140	532a	32	1090	2120a	1120
Ce-141	3560	--	6040a	--	10600
Ce-144	2690	473	5000a	6060	8370
Pb-210	< 0.3	0.76	<0.8	0.79b	<1
Pu-238	0.21a	0.07a	0.29a	0.30a	0.46a
Pu-239	1.0	0.25	1.7a	2.2	2.6

Atmospheric Radioactivity Data, January 1970

Latitude	30-35°N	9-10°N	5-10°N	5-10°N	5-10°N
Longitude	103-106°W	79°W	79°W	79°W	79°W
Date	5	20	20	5	19
Altitude (km)	19.8	12.2	12.2	15.2	15.2
Tropopause Ht. (km)	9.4-12.8	16.2	16.2	15.5-15.8	16.2
Isotopes					
Be-7	9940	<180	<110	620	170c
Mn-54	35	< 2	< 1	< 0.7	< 2
Fe-55	108b	<10	--	< 5	--
Sr-89	6630	6.3a	8.9a	3.2a	0.87c
Sr-90	181	0.75b	0.89	0.38a	<0.6
Zr-95	11500	15	18	5.9a	3.4a
Cs-137	266	1.4a	2.1a	0.54a	< 1
Ba-140	4290c	NR	NR	NR	NR
Ce-141	--	--	5.1c	--	820
Ce-144	6650	2.7a	14	3.8a	3.8a
Pb-210	0.35b	0.27c	<0.4	0.23b	<0.9
Pu-238	0.31	< 0.02	0.02b	< 0.01	NR
Pu-239	1.9	0.01c	0.01a	< 0.01	NR

Atmospheric Radioactivity Data, January 1970

Latitude	5-10°N	5-6°N	5-10°N	5-10°N	34-39°S
Longitude	79°W	79°W	79°W	79°W	68°W
Date	5	5	19	13	6
Altitude (km)	18.3	18.3	19.2	19.5	12.2
Tropopause Ht. (km)	15.5-15.8	15.5-15.8	16.2	15.8	10.7-11.6

Isotopes

Be-7	5810a	24400	1200	3820a	1070
Mn-54	4.4b	14	2.6a	7.3a	< 0.8
Fe-55	--	< 50	13	--	< 5
Sr-89	1210	1830	143	1230	0.75a
Sr-90	55a	76	17	80a	1.1
Zr-95	2320	3170	255	2250	4.8
Cs-137	88	120	28	119	1.8
Ba-140	NR	NR	NR	NR	NR
Ce-141	1310	--	--	< 8	--
Ce-144	1470	1880	295	1560	15
Pb-210	< 1	1.6b	0.50a	< 1	0.67a
Pu-238	0.14a	0.26b	0.10a	0.20a	0.01d
Pu-239	0.53	0.82a	0.19	0.77	0.01b

Atmospheric Radioactivity Data, January 1970

Latitude	40-45°S	34-40°S	40-45°S	35-40°S	40-45°S
Longitude	67°W	68°W	67°W	68°W	67°W
Date	19	5	20	12	19
Altitude (km)	12.2	15.2	15.2	18.3	18.3
Tropopause Ht. (km)	15.8	10.7-11.7	14.0	10.7-11.3	15.8
Isotopes					
Be-7	1320a	14000	3190	2480	13500
Mn-54	3.6a	6.0b	1.5a	8.5a	7.1a
Fe-55	--	--	< 6	39a	--
Sr-89	1.4d	2700	8.3	17b	16
Sr-90	13	138a	7.9	80	63
Zr-95	11	5360	18	49	67
Cs-137	19	218	13	67	92
Ba-140	NR	NR	NR	NR	NR
Ce-141	< 2	3230	--	--	28d
Ce-144	138	3090	93	530	658
Pb-210	< 0.2	< 0.6	0.77a	0.86a	< 0.8
Pu-238	0.11a	0.47a	0.08a	0.35a	0.52a
Pu-239	0.13	1.6	0.09a	0.46a	0.62

Atmospheric Radioactivity Data, January 1970

Latitude	40-45°S	40-45°S
Longitude	67°W	67°W
Date	12	12
Altitude (km)	18.9	19.2
Tropopause Ht. (km)	10.7-11.3	10.7-11.3
Isotopes		
Be-7	7440a	8380
Mn-54	9.3a	15a
Fe-55	--	48a
Sr-89	10c	17b
Sr-90	72	78
Zr-95	63	72
Cs-137	101	111
Ba-140	NR	NR
Ce-141	31d	--
Ce-144	758	889
Pb-210	< 4	0.50b
Pu-238	0.62a	0.68a
Pu-239	0.64a	0.81

PART IV

RECENT PUBLICATIONS RELATED TO RADIONUCLIDE STUDIES

Recent Publications Related to Radionuclide Studies

- Beasley, T.M. and E. E. Held
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