BNWL-1551
Part 2
UC-48

PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1970
TO THE USAEC DIVISION
OF BIOLOGY AND MEDICINE

VOLUME II: PHYSICAL SCIENCES
PART 2. RADIOLOGICAL SCIENCES

March 1971

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DEVELOPMENT REPORT

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BATTELLE MEMORIAL INSTITUTE

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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1970
to the
USAEC Division of Biology and Medicine
VOLUME II: PHYSICAL SCIENCES
Part 2. Radiological Sciences

By
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and
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Environmental and Life Sciences Division

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FOREWORD

This report to the USAEC Division of Biology and Medicine covers work done in the fields of environmental and radiological chemistry, radiation chemistry, radiation instrumentation, radiation dosimetry, and radiation and health physics. It continues a series of reports initiated in 1959. Special effort has been made to make the report a brief and concise technical summary of the accomplishments. For details of the work, references are given to the pertinent publications and presentations resulting from the studies.

Reports previously issued in this series are as follows:

HW-63824	July 1, 1959 - December 31, 1959
HW-70050	Period Ending December 31, 1960
HW-73337	Period Ending December 31, 1961
HW-77609	Period Ending December 31, 1962
HW-81746	Period Ending December 31, 1963
BNWL-36 III	Period Ending December 31, 1964
BNWL-235 Part 2	Period Ending December 31, 1965
BNWL-481 Part 2	Period Ending December 31, 1966
BNWL-715 Part 2	Period Ending December 31, 1967
BNWL-1051 Part 2	Period Ending December 31, 1968
BNWL-1307 Part 2	Period Ending December 31, 1969

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RADIOLOGICAL CHEMISTRY

INHALATION HAZARDS TO URANIUM MINERS

The high incidence of lung cancer and other respiratory ailments in uranium miners requires the accurate characterization of the radionuclide, elemental and organic content of the uranium mine atmosphere and their correlations to lung exposure.

Characterization and Procedure
Development for the Measurement of
the Uranium Mine Atmospheric Con-
stituents and Their Retention in
Human Lungs

J. A. Cooper, H. G. Rieck, Jr.,
P. O. Jackson, L. A. Rancitelli,
W. A. Haller, R. W. Perkins and
A. D. Renzetti, Jr.*

Atmospheric contaminants encountered in underground uranium mines are known to present serious occupational health hazards. However,

evaluation of the detrimental effects of the gaseous and aerosol components requires that their composition be known, and for aerosol particles, that their composition as a function of particle size be known. A sensitive research instrument based on simultaneous α - and γ -ray spectroscopy has been developed which permits the continuous measurement of radon daughter concentrations in uranium mine and experimental animal chamber atmospheres as a function of particle size. The integrated radon daughter exposure is subsequently related to the integrated elemental content of the mine atmosphere as a function of

* The University of Utah Medical Center, Salt Lake City, Utah

particle size by newly developed neutron activation and radioisotopic X-ray fluorescence analysis procedures. Short successive counting intervals provide a continuous record of the atmospheric radon daughter concentrations for the particle size ranges being selected by the cascade impactor's air inlet. The subsequent analyses of the impactor stages and of the air filter which is viewed by the α - and γ -ray spectrometers yield elemental, radiochemical and organic constituent concentrations of the atmosphere being studied. Data on uranium chain members and other possible carcinogenic constituents whose individual or synergistic effects may promote lung damage are thus obtained. The continuous monitoring instrument is reasonably sensitive to minor changes in the radon daughter concentration which do occur in mines and animal test chambers.

Measurement of the low activity levels of long-lived uranium chain members in mine atmosphere and tissue samples from animal exposure studies requires high sensitivity technology and usually necessitates prior chemical purification. A relatively simple technique with high sensitivity has been developed for counting estimated radon in radium analysis. It consists of counting a purified radium solution which has been sealed for 30 days in a small evacuated vial whose walls are painted with a zinc sulfide scintillator-mineral oil mixture. An average radiochemical recovery of $94 \pm 4\%$ has been achieved in radium separations from dissolved animal tissue with a counting efficiency of

1.6 counts per disintegration and a background (excluding blank which was 0.4 count per minute) of 0.03 count per minute. The technique has been applied to hamster tissue samples in which U, ^{230}Th , ^{210}Pb and ^{210}Po were also measured. The concentrations of ^{226}Ra measured in tissue samples of about one-half gram ranged from 3 to 11 disintegrations per minute (dpm). A computer procedure has been developed for the evaluation of counting errors associated with the measurement of the daughter-parent couple of ^{210}Po and ^{210}Pb when using ^{208}Po as an internal standard which allows rapid evaluation of the precision as well as recovery and counter efficiency.

Lung tissue samples from individuals with no mining history are being analyzed by neutron activation analysis to provide a base line comparison of individuals with mining experience. The elemental composition of a large number of lung tissue samples from normal individuals has thus been analyzed and compared for elemental correlations assuming a bilateral normal distribution. Many significant element correlations have been demonstrated at 1% confidence level in a group of 35 lung tissue samples from normal individuals. Some of the more important and surprising correlations are those between Se and Co, Ag and Co, and Hg and Sc in addition to the well-known correlation such as those between Pb and K. The observed positive correlation between K and Na was surprising in the light of previously demonstrated different biological properties.

Neg 705548-3

Schematic Representation of the
Detector-Filter Chamber Used
for the Continuous Measurement
of Radon Daughters in Uranium
Mines and Animal Test Chambers

Neg 700601-2

Photograph of the Disassembled
Detector-Filter Chamber Used
for the Continuous Measurement
of Radon Daughters in Uranium
Mines and Animal Test Chambers

Elemental Correlation Matrix for Lung Tissue Samples
from Individuals with No Mining History

(Sample Size = 35)

	Na	K	Rb	Cs	Br	Zn	Fe	Co	Cr	Se	Ag	Hg	Sb	Sc
Na	1.000	0.434**	-0.183	0.101	0.207	0.116	-0.358	0.214	-0.294	0.589**	0.042	-0.123	-0.366*	-0.246
K		1.000	0.456**	-0.335*	-0.434**	-0.239	-0.120	-0.286	-0.046	0.274	-0.206	-0.264	-0.134	-0.233
Rb			1.000	-0.362*	-0.631**	-0.152	0.176	-0.470**	-0.103	-0.101	-0.268	-0.257	0.308	-0.228
Cs				1.000	0.103	0.435**	-0.087	0.253	-0.107	0.095	0.048	0.663**	-0.102	0.309
Br					1.000	0.275	-0.131	0.565**	-0.043	0.246	0.498**	-0.107	-0.155	-0.080
Zn						1.000	-0.116	0.136	0.042	0.289	0.134	0.138	-0.067	0.109
Fe							1.000	0.191	0.446**	-0.104	0.041	-0.253	0.707**	0.317
Co								1.000	0.019	0.449**	0.564**	-0.160	0.270	0.055
Cr									1.000	0.142	-0.075	-0.016	-0.006	0.782**
Se										1.000	0.206	-0.105	-0.145	0.095
Ag											1.000	-0.069	0.096	-0.034
Hg												1.000	-0.350	0.492**
Sb													1.000	-0.075
Sc														1.000

* Significant at the 0.05 level of significance

** Significant at the 0.01 level of significance

RATES OF DEPOSITION OF AIRBORNE RADIONUCLIDES
IN THE ALASKAN ENVIRONMENT

The atmospheric deposition rates, routes, biological uptake rates and concentrations of fallout radionuclides and their stable element counterparts are being investigated in the terrestrial and marine environments, with emphasis on the food chains that consistently lead to high radionuclide concentrations.

Deposition of Fallout, Cosmic and
Natural Radioelements in Alaska

C. E. Jenkins

Eighteen radionuclides are being traced through the following three ecosystems in Alaska: the marine system centered at Kotzebue, the Arctic centered at Anaktuvuk Pass and the forest centered in the Kenai Peninsula near Anchorage. Continuing studies of the marine and Arctic environments have defined many of the crucial radioelements in these ecosystems. However, knowledge of the relative behavior of radioelements in a forest ecosystem which covers most of Alaska is largely undeveloped. Thus, samples were taken to elucidate the behavior of radioelements in the Chugach Forest on the Kenai Peninsula and comparisons were made with the marine and Arctic ecosystems. Preliminary estimates indicate that cos-

mogenic radionuclides, ^7Be and ^{22}Na are two- to fourfold higher in lichens and mosses at Anaktuvuk than at Kenai during the summer months. Shorter-lived fallout radioelements appear to be in higher concentrations in the Kenai Peninsula than in the Anaktuvuk region while the reverse seems to be true for the longer-lived fallout radioelements. Olympic Peninsula fallout radioelemental concentrations are higher than either Alaskan site. Direct comparisons are difficult because the Olympic Peninsula samples were obtained during the spring maximum fallout input whereas the Alaskan samples were taken near the seasonal low. Concentrations of ^{238}U and ^{232}Th decay chain radionuclides appear to be disproportionately high throughout all Kenai samples considering the total rainfall (50 inches) as compared to the Olympic Peninsula (140 inches).

Comparison of Radionuclides in Alaska and Washington - 1970

(dpm/kg wet wt)

Radio-nuclide	Anaktuvuk Pass, AK, (Mixed) Lichen 8/70	Anaktuvuk Pass, AK, Grnd. Moss 8/70	Kenai Pen., AK, Grnd. Moss 8/70	Olympic Pen., WA, Hanging Moss 3/70	Kenai Pen., AK, Log Top Moss 8/70	Olympic Pen., WA, Log Top Moss 3/70
⁷ Be	14,000		2,340	5,220		3,660
²² Na	6.1	3.14	1.82	4.95	2.75	6.1
⁴⁰ K	1,140	1,170	1,380	8,710	3,100	7,240
⁵⁴ Mn	169	115	97.9	424	132	904
⁶⁰ Co	45.9	44.5	28.0	106	53.7	224
⁶⁵ Zn	258	130	10.6	389	29.7	916
⁹⁵ Zr-Nb	2,910	1,800	3,780	8,130	3,750	6,640
¹⁰³ Ru		69.4	246	4,580		2,000
¹⁰⁶ Ru	2,250	1,840	3,070	5,110	3,120	4,610
¹²⁵ Sb	367	1,560	977	4,110	1,830	8,080
¹³⁷ Cs	22,200	13,400	8,200	31,200	12,800	68,300
¹⁴¹ Ce						
¹⁴⁴ Ce-Pr	7,390	4,890	8,340	16,500	8,350	18,500
¹⁵⁵ Eu						
²⁰⁸ Tl	21.6	17.2	19.2	41.4	87.5	39.5
²¹⁴ Bi	55	76.4	23.3	136	84.7	110

¹⁰⁸Ag in Terrestrial and Marine
Organisms

C. E. Jenkins and J. A. Cooper

The recent development of a dual Ge(Li) diode system has allowed positive analysis of ^{108m}Ag. The first tentative measurements were made in 1967-68 using anticoincident shielded NaI(Tl) multidimensional spectrometers or large volume Ge(Li) detectors. Gamma-ray spectra from a single Ge(Li) diode had only the 723 keV peak from

the triple coincident gamma-ray decay of ^{108m}Ag due to the Compton backscatter from high levels of ¹³⁷Cs. Silver identification from NaI(Tl) multidimensional spectra was complicated by low levels of ^{108m}Ag relative to high levels of other radioelements which backscattered into the coincidence regions of interest. By separating anticoincident and coincident peaks using the dual high-resolution Ge(Li) diodes, the triple coincidence peaks (434, 615 and 723 keV) of

^{108m}Ag were easily identified and measured. Earlier investigations have shown that silver concentrates primarily in liver tissues and is associated with blood and bone marrow. The ratio of ^{110m}Ag in seawater to that in marine organisms indicates concentration factors of 10^4 to 10^6 .

Terrestrial herbivores such as elk, deer, and caribou concentrate silver in the liver by one to two orders of magnitude. In the case of caribou these levels are elevated by the efficient long-term retention of fallout elements by lichen and thus are easily detectable.

The ratio of $^{108m}\text{Ag}/^{110m}\text{Ag}$ and the Ag concentrations provide a means of estimating the past production of

^{108m}Ag as well as an estimation of the $^{107}\text{Ag}(n,\gamma)^{108m}\text{Ag}$ cross section. Based on the 1967 ratio and using September 1962 as the production maximum, it appears that there was approximately 250 times more ^{110m}Ag produced than ^{108}Ag ; thus, the $^{107}\text{Ag}(n,\gamma)$ cross section appears to be between 0.006 and 0.01 barn.

Since Ag appears to have an increasing role in weather modification and is a highly toxic element to many life forms, the ecological role of Ag becomes increasingly important. In view of this and the increasing levels of Ag introduced by industrial waste, more information concerning the ecological behavior of fallout silver is imperative.

Neg 704620-5

Anticoincidence-Shielded Dual Ge(Li) Gamma-Ray Spectra
of Caribou Liver

Radionuclide Concentrations in North
Pacific Salmon

C. E. Jenkins

Fallout ^{55}Fe is rapidly assimilated by the oceanic biomass, and has a turnover time between 9 and 10 months which is similar to the stratospheric residence half time. Fallout radioelements such as ^{55}Fe and ^{54}Mn are tenfold higher in northern (Alaskan) compared to southern (Washington-Oregon) waters. An additional radionuclide source is the Hanford Plant from which radioelements such as ^{65}Zn and ^{51}Cr are introduced into the oceans via the Columbia River.

Salmon ^{55}Fe concentrations show distinct plateaus at different latitudes which suggest distinct feeding practices. Iron-55 concentrations decline ten- to thirtyfold from northern to southern latitudes depending on the chum salmon run. Immature salmon show depressed ^{55}Fe levels which reflect the higher stable Fe concentration of river waters in which they recently resided. Northern chum salmon are three- to tenfold higher in ^{55}Fe than Anaktuvuk caribou and 300 to 1000 times higher than Richland cattle. Thus salmon-eating Eskimos have 20- and 100-fold higher body burdens than

caribou-eating Eskimos and Richland residents, respectively. However, ^{55}Fe concentrations are declining at a more rapid rate (2.5 times) in the oceans than in the terrestrial environments which is reflected by a reduced body burden difference between salmon-eating Eskimos and the rest of the populace.

Zinc-65 concentrations in mature salmon are maximum near the mouth of the Columbia River. Immature king salmon ^{65}Zn concentrations are maximum north of the Columbia near Westport, Washington suggesting that these salmon are from the Columbia River area and are on a northerly migratory pattern. Zinc-65 levels decline in immature king salmon taken at Neah Bay, Washington and suggest that these fish are from a more northerly river than the Columbia. Zinc-65 concentrations were observed at all latitudes which suggests that some ^{65}Zn contribution is from fallout. Cesium-137 concentrations peak slightly at the mid-latitudes (45° to 50° N) with the king salmon having the highest concentrations. The relatively constant ^{137}Cs level in salmon is indicative of the existence of a large well-mixed reservoir of this long-lived radioelement in the ocean.

Distribution of ^{65}Zn in North American Salmon in 1968 Distribution of ^{55}Fe in North American Salmon in 1968

Tracer Element Correlations in Pacific Salmon

L. A. Rancitelli

The trace element content of Pacific salmon has been extensively characterized in an attempt to understand the pathways and biological uptake of fallout and manmade radionuclides in the marine environment. Relationships between the alkali metals K, Rb and Cs were detected which indicated that the radiologically important element Cs will be preferentially assimilated in the food chain. The relation of Cs to K and Rb was readily obtained since the simple monovalent chemistry of the alkali metals permitted a direct comparison of the relative concentrations in the organisms with those in seawater. Unfortunately, the biochemistry of other elements considered in the study of Pacific salmon is too complex to permit a valid intercomparison with seawater. Since the study involved the simultaneous determination of 14 elements in each tissue specimen, a statistical analysis of the data for trace element correlations was made in an attempt to reveal similar biochemical behaviors.

A normal bivariate distribution was assumed for pairs of elements, and the null hypothesis that the correlation coefficient is different from zero was

tested. The statistical analysis revealed several interesting interelemental relationships. The correlation coefficient for the Se-Ag content of liver of 0.823 is different from zero at a significant level of one part per million for the number of samples considered, indicating only an extremely remote possibility of a random correlation. The correlation of Se and Ag in the liver may reflect the fact that Ag is complexed by biochemical sulfur groups with which Se is closely related. The interesting biochemical relationship of Se and Ag also suggests that the turnover of ^{110m}Ag may be related to the Se and/or S cycle. A number of other radiologically important elements were found to have correlations at the 0.01 level of significance in salmon liver and muscle. In addition to the Rb-K relationship noted previously, these include Fe-Na, Zn-K, Fe-Cs, Zn-Br, Br-Co, Co-Zn, and Co-Fe in muscle and Fe-Na, and Ag-Na in liver. While the biochemical interpretation of these correlations has not been fully explored as yet, it appears that they will prove valuable in developing models of radionuclide uptake based upon specific activities. This should be particularly true of pairs of elements such as Ag-Cs in the liver which have a large negative correlation coefficient indicating an inverse relationship between their correlations.

Liver Correlation Matrix

Element	Na	K	Rb	Cs	Br	Zn	Fe	Co	Ag	Se
Na	1.000	-0.005	0.075	-0.045	0.111	-0.009	0.438*	-0.197	0.454**	0.257
K		1.000	0.407*	0.296	-0.072	-0.097	-0.284	0.049	-0.327	-0.239
Rb			1.000	0.182	0.089	-0.032	0.130	0.052	-0.310	-0.495**
Cs				1.000	0.163	-0.023	0.222	0.018	-0.572**	-0.575**
Br					1.000	0.130	0.418*	0.364*	-0.135	-0.187
Zn						1.000	0.303	-0.015	-0.205	-0.179
Fe							1.000	0.198	0.224	-0.041
Co								1.000	-0.295	-0.294
Ag									1.000	0.822**
Se										1.000

* Significant at the 0.05 level of significance.

** Significant at the 0.01 level of significance.

PHYSICAL AND RADIOLOGICAL CHEMISTRY OF OCEAN SOLUTIONS

The concentrations, distributions and behavior of the oceans' trace elements and natural and artificial radio-nuclides are being investigated. The characterization of these constituents in the marine environment is contributing to a better understanding of ocean mixing, vertical transport, and the biogeochemical and physical processes affecting the apportionment and fate of the oceans' radioactive and stable elements.

Silver Concentrations in Marine
Organisms and Seawater

D. E. Robertson

The increasing use of silver iodide as a cloud-seeding agent during extensive, worldwide weather modification studies has prompted a growing concern for the behavior of silver in the marine environment. Silver ion is one of the most toxic heavy metals to aquatic microorganisms and animals, and previous studies have shown that levels as low as 1 to 10 ppb are lethal to some organisms. Since the concentrations of silver in seawater and marine organisms are not well known, this study was initiated to determine present levels of silver in the ocean and in a wide variety of marine organisms, so that more intelligent predictions of the impact of using silver iodide in weather modification studies could be evaluated.

Silver concentrations were determined in Northeast Pacific and Northwest Atlantic marine organisms, and in seawater samples collected in a Northeast Pacific Ocean (28°29' N Lat., 121°38' W Long.) depth profile. In-

strumental neutron activation analysis, utilizing high sensitivity multidimensional gamma-ray spectrometers, was employed in the measurements. Silver was determined directly in freeze-dried tissues of marine organisms, and a preconcentration technique involving cocrystallization of silver from seawater with thionalid (2 mercapto-N-2-naphthylacetamide) was used to isolate silver from 1 liter samples of seawater. The principal conclusions regarding silver in the marine environment are as follows:

- Many marine organism tissues, particularly albacore liver and the internal organs of squid, shrimp and several lantern and pelagic fishes, contained relatively high concentrations (10 to 4900 ppb) of silver. Muscle tissues normally contained much lower concentrations of silver than internal organ tissues. Silver appears to be concentrated in the liver.
- Silver is accumulated from seawater by marine organisms by factors frequently ranging from four to five orders of magnitude.

- The silver concentration in the Pacific Ocean depth profile averaged 0.0078 $\mu\text{g Ag/liter}$, and ranged from 0.0023 to 0.019 $\mu\text{g Ag/liter}$. These values are approximately 1/20 to 1/50 of previous estimates of silver in the Pacific Ocean.
- The silver distribution with depth, except for an anomalously high value at 50 meters, exhibited a maximum concentration at a depth of 700 meters, which correlated with the oxygen minimum layer in this profile. The high silver concentration at the oxygen minimum layer is explained by the regeneration and accumulation of silver at this depth by the oxidation and decomposition of sinking detritus and dead organisms containing relatively high silver levels.
- The higher silver levels in the Atlantic Ocean zooplankton and

Silver Concentrations in Marine Organisms

<u>Organism</u>	<u>Silver Concentration, ppb wet weight</u>	<u>Concentration Factor*</u>
<u>Northeast Pacific Ocean</u>		
Mixed Zooplankton	<1	<130
Albacore Flesh	2.8 - 8.8	330 - 1100
Albacore Liver	82 - 770	10,000 - 100,000
Squid - Flesh	46	5900
Squid - Internal Organs	250	33,000
Shrimp - Tail	7.3	950
Shrimp - Whole	10-240	1300 - 31,000
Shrimp - Internal Organs	258	33,000
<u>Lantern Fishes</u>		
Flesh	<0.04 - <4	<5 - <50
Whole	2 - 74	250 - 9500
Internal Organs	25 - 521	3300 - 67,000
<u>Pelagic Fishes</u>		
Flesh	0.5 - 8.8	65 - 1100
Whole	1.5 - 5.5	190 - 720
Internal Organs	1.0 - 68	130 - 8700
<u>Tropical Northwest Atlantic Ocean</u>		
Mixed Zooplankton	17	2,200
Squid - Flesh	5	640
Squid - Liver	4900	640,000
Squid - Ink Sac	1900	240,000
Squid - Gonad	1300	170,000
Squid - Heart	510	67,000

* The concentration factor is defined as the silver concentration in the wet weight organisms divided by the average silver concentration of Pacific Ocean seawater (0.0078 $\mu\text{g Ag/liter}$).

squid, relative to Pacific Ocean species, may be an indication of biological uptake of silver introduced to the Caribbean Sea and Western Atlantic Ocean by weather modification activities involving silver iodide seeding in these areas.

- In view of the biological activity of silver in the marine environment and the fact that silver is present in the oceans at much lower concentrations than previously believed, a re-evaluation of the ecological effects of silver added to the oceans during weather modification studies is necessary.

Silver Concentrations of Pacific
Ocean Seawater 28°29' N Lat.
121°38' W Long.
September, 1969

Depth, meters	Silver Concentration, µg/liter
10	0.0032
50	0.0190
200	0.0023
350	0.0041
550	0.0056
700	0.0120
1,000	0.0056
2,000	0.0095
3,000	0.0100
4,000	0.0063
Avg. Conc.	0.0078

Application of Instrumental Neutron
Activation Analysis in the Study of
Marine Sediment Interstitial Waters
D. E. Robertson and J. L. Bischoff*

Recently, it has been recognized that the interstitial waters (pore fluids) contained in marine sediments act as a significant reservoir and participant in the marine geochemical cycle by redistributing elements through the pore fluid from sediments back to seawater. A classic example of this phenomenon is the dissolution of radium from deep-sea sediments and its subsequent penetration into the bottom waters of the oceans. It is well recognized that pore fluids are generally slightly enriched with some major constituents of seawater (K^+ , SiO_2 , Li^+ , PO_4^{3-} , B) and depleted in others (SO_4^{2-} , Mg^{++} , Ca^{++}). However, very little information is available concerning the concentrations and behavior of trace elements in pore fluids. To determine the significance of this source of trace elements to deep waters of the ocean, a cooperative study was started to characterize the trace element distribution in pore fluids. To demonstrate the applicability of instrumental neutron activation analysis to this problem, three samples of pore water (supplied by Dr. Bischoff) were neutron activated and counted directly on a multidimensional gamma-ray spectrometer

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and a Ge(Li) diode detector after an appropriate decay period. The excellent sensitivity of this method permitted the measurement of Sc, Co, Ag, Sb, Cs, U, Zn, Fe and Cu in only 1.9 to 2.6 ml of pore water. It is evident that these pore waters were greatly enriched with most trace elements, relative to their concentrations in seawater. Uranium exhibited the most diverse behavior, being depleted by about a factor of 10 in Sample 42, enriched by about a factor of 10 in Sample 25 and remaining just slightly under the normal seawater concentration in Sample 11. In addition to its high uranium content, Sample 25 also contained an extremely high silver concentration. Zinc and cobalt were greatly enriched in all of the pore waters, while Sc and Sb exhibited lesser dissolution from the

sediments. The iron values were surprisingly low, but any iron which may originally have been present was probably lost by adsorption onto the polyethylene container walls. A major problem in studying the trace element constituents of pore waters is contamination of the sample during manipulation. The apparatus used for squeezing pore waters from the sediments used in this study was constructed of stainless steel and minor constituents of the steel could have contaminated the pore water during its extraction from the sediments. However, contamination is an unlikely explanation for the very unusual uranium concentrations in these pore waters, and it appears that some elements other than radium may enter bottom waters by dissolution of certain sediments.

Trace Element Concentrations in Pore Waters

Sample	Identification	Chlorinity, %	Sample Wt, g	Concentration, $\mu\text{g/liter}$								
				Sc	Co	Ag	Sb	Cs	U	Zn	Fe	Cr
A-II-54-11	Pilot	19.33	2.61	0.008	1.8	~0.09	0.46	0.39	2.4	140	<30	~0.7
A-II-54-25	Top, #2	19.489	2.03	0.001	0.52	1.6	1.5	0.26	40.4	190	<20	--
A-II-54-42	Top, #2	19.67	1.88	0.008	4.7	~0.07	1.8	0.33	0.36	320	48	--

Ratio of: $\frac{\text{Trace Element Concentration in Pore Water}}{\text{Trace Element Concentration in Typical Oceanic Water}}$

	Sc	Co	Ag	Sb	Cs	U	Zn	Fe	Cr
A-II-54-11	13	60	10	2.1	1.3	0.73	47	<6	3.5
A-II-54-25	2	17	200	6.8	0.87	12.0	63	<4	--
A-II-54-42	13	160	10	8.2	1.1	0.11	107	10	--

Trace Element Concentrations in
Coastal Waters Surrounding the
British Isles

D. E. Robertson

Comparative studies with the Fisheries Radiological Laboratory in Lowestoft, Great Britain are continuing to characterize the distribution of trace elements in coastal waters surrounding the British Isles. These waters are particularly interesting for two reasons: (1) fission and activation products are discharged into the Irish Sea from the Windscale spent-fuel reprocessing factory and (2) many of the early analyses of trace elements in seawater reported in the literature were determined in samples from these waters, which serve as a base line for comparing later measurements. In November 1969 and May 1970 two cruises were conducted during which surface seawater samples were collected for the determination of trace elements by instrumental neutron activation analysis.

British Isles Coastal Seawater
Sampling Stations

The results of these analyses are compared with the average concentrations of trace elements measured in a similar investigation conducted in 1968. Except for several samples collected in shallow waters during stormy weather, the trace element distributions in these coastal waters were surprisingly similar to those in the oceanic waters of the Atlantic. Only the iron concentrations were significantly higher than in open ocean waters, being highly variable as a result of terrestrial material undoubtedly present in the water at several stations. Cesium, uranium and antimony were quite homogeneously distributed in the waters surrounding the British Isles at concentrations averaging 0.30, 3.3 and 0.29 $\mu\text{g/liter}$, respectively. The Zn and Co concentrations were much lower than would have been predicted for such nearshore waters and, except for Stations 10 and 11, are typical of oceanic concentrations. The scandium concentrations are nearly ten times higher than oceanic waters, which is the result of terrestrial material or suspended sediments present in the water. The average concentrations of Sc, Co, Zn and Fe measured in this study vary significantly from their average concentrations determined in 1968. However, due to the variable input of terrestrial material into these nearshore waters as a function of time, weather, coastal currents, etc., it is difficult to establish accurate average concentrations for Sc, Co, Zn and Fe. These studies have substantially contributed to a better understanding of the composition and

distribution of trace constituents in the coastal waters surrounding the British Isles, and are being used by the Fisheries Radiobiological Labo-

ratory to establish present specific activities of the radioactive nuclides of many of these elements discharged into the Irish Sea at Windscale.

Trace Elements in British Isles Coastal Surface Waters
(Concentrations in $\mu\text{g/liter}$)

<u>Station</u>	<u>Depth, meters</u>	<u>Sea State</u>	<u>Sc</u>	<u>Co</u>	<u>Sb</u>	<u>Cs</u>	<u>U</u>	<u>Zn</u>	<u>Fe</u>
1	37	1	0.006	0.046	0.29	0.29	3.2	1.7	19
2	48	3	0.009	0.013	0.23	0.29	3.4	1.0	6.0
3	110	3	0.006	0.010	0.24	0.30	3.4	1.2	16
4	64	3	0.002	0.017	0.27	0.28	3.3	3.3	17
5	64	3	0.008	0.018	0.26	0.28	3.2	0.93	4.8
6	60	4	0.007	0.010	0.25	0.30	3.5	0.44	11
7	137	2	0.006	0.026	0.23	0.29	3.0	2.9	41
8	86	3	0.002	0.018	0.23	0.29	3.2	1.9	18
9	29	3	0.007	0.037	0.33	0.32	3.4	6.8	47
10	55	5	0.018	0.037	0.56	0.32	3.3	188*	77*
11	26	4	0.052*	0.133*	0.29	0.35	3.5	5.8	262*
Avg. Conc.			0.007	0.022	0.29	0.30	3.3	2.6	20
Avg. Conc. in 1968			0.002	0.040	--	0.27	3.0	6.5	8.8

Samples 1 to 6 were collected during May, 1970 and samples 7 to 11 were collected during November, 1969.

* Samples suspected to be contaminated by terrestrial material; these values not included in average concentrations.

Behavior of Trace Constituents of
Seawater in Contact with Various
Pelagic Sediments

D. E. Robertson and E. K. Duursma*

A knowledge of the interactions of suspended sediments with the radio-nuclides deposited in the ocean is basic to an understanding of the behavior and fate of the radioactivity entering the marine environment. Previous studies by Duursma and others have shown that deep sea sediments can be effective adsorbents for removing several fission product radio-nuclides from solutions of seawater. It has been proposed that these sorptive properties of pelagic sediments be practically applied for the decontamination of ocean areas polluted by radioactive materials.

Essentially all of the previous studies concerning the sorptive properties of sediments involved the use of radioactive tracers added to suspensions of sediments in seawater, and consequently very little is known about the sorption behavior of the natural trace element constituents normally present in the oceans.

In a cooperative study an experiment was devised to study the behavior of the natural trace constituents in seawater during contact with various types of pelagic sediments. Five-hundred-milliliter samples of Mediterranean Sea water were contacted with about 6-gram samples of wet

pelagic sediments collected from the Mediterranean, Pacific, Antarctic, Bahamas and the Var River (Nice). The suspensions were kept at 4 °C for 3 weeks in the dark and shaken for 1 hour each day. After 3 weeks the samples were centrifuged and aliquots of seawater were siphoned off for trace element analysis. Samples of the original and the contacted sediments and the original untreated seawater were also taken for analysis. Twenty-five-milliliter aliquots of the original and treated seawater and about 20-milliliter samples of sediments were analyzed by instrumental neutron activation analysis utilizing multidimensional gamma-ray spectrometry and Ge(Li) diode spectrometry. Quantitative results were obtained for the seawater analysis by a comparator method using standards of all the elements measured, except Sr and Rb. Therefore only relative concentrations (compared to the original seawater) of Sr and Rb were determined. The sediment analyses were quantified using both the comparator and the flux method with the U.S. Geological Survey standard rocks W-1 and G-2 and dilute Co solutions as the standards.

The results of the analyses were quite unexpected. Instead of the natural trace elements in the seawater being depleted from solution by adsorption on the sediments, the reverse apparently occurred, and the concentrations of these elements in the seawater, except for Sr, Rb and in three samples for Zn, actually increased. Either partial dissolution of the

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sediments had occurred to increase the concentrations in the seawater or colloidal, noncentrifugable particles of sediment had remained in solution after the contacting treatment. The elemental concentrations of the sediments were so high, relative to their concentrations in the seawater, that the precision of the sediment analysis was not great enough to detect small losses by dissolution of trace elements from the sediments. However, in all cases where the trace element concentrations in the sediments were high, a corresponding increase was observed in the seawater. The anomalous trace element concentrations in the treated Sediment 5 cannot be explained at present, since the treated

Seawater Sample 5 showed no such anomaly which could account for the observed concentrations in the sediment. The apparent disagreement concerning the sorptive properties of sediments in this study, compared with earlier experiments utilizing radioactive tracers, is perplexing, and points out the need for further studies designed to elucidate the sorption behavior of both the stable, natural trace elements and radioactive tracers. This present study would indicate that pelagic sediments would not be effective in decontaminating areas of the ocean polluted with radioactive materials, a conclusion contrary to that derived from tracer experiments.

Trace Element Concentrations in Seawater and Sediments
Before and After Contacting for 3 Weeks

Seawater	Concentrations in ppb (ug/liter)							Sr-relative to Original	Rb-relative to Original
	Zn	Fe	U	Cs	Sb	Co	Cs		
1. Original	26	6	3.8	0.33	0.26	0.023	0.0023	--	--
2. Contacted with Sediment 2	5	51	5.5	0.37	0.43	0.046	0.0073	1.00	0.99
3. Contacted with Sediment 3	5	86	10.4	0.54	0.36	0.128	0.0165	1.05	0.96
4. Contacted with Sediment 4	71	26	5.3	0.38	0.31	0.069	0.0063	1.14	1.05
5. Contacted with Sediment 5	23	24	4.6	0.37	0.38	0.083	0.0070	1.00	0.91
6. Contacted with Sediment 6	33	33	5.4	0.51	0.37	0.032	0.0038	1.16	1.04

Sediments	Concentrations in ppm										
	Sr	Cr	Sc	Co	Ce	Eu	Sb	Cs	Hf	Th	Fe
Original-2 (Mediterranean)	1700	96	10	14	42	0.94	0.12	5.5	1.9	8.8	3.2
Treated-2 (Mediterranean)	<1000	130	12	19	49	1.7	0.17	6.4	2.1	10	4.0
Original-3 (Pacific)	< 800	87	21	130	87	2.7	0.33	8.4	4.0	15	5.8
Treated-3 (Pacific)	< 800	65	21	104	83	4.4	0.34	8.4	5.0	18	2.8
Original-4 (Antarctic)	3400	4.8	1.9	4.8	5.6	0.54	0.018	0.39	0.33	1.3	0.38
Treated (Antarctic)	3500	3.6	1.6	4.2	4.0	0.36	0.024	0.33	0.13	0.96	0.33
Original-5 (Bahamas)	3100	38	7.9	20	3.5	0.87	0.053	2.3	1.7	5.2	2.5
Treated-5 (Bahamas)	13000	7.5	0.25	0.45	1.9	0.098	0.018	0.48	0.31	0.16	0.069
Original-6 [Var River (Nice)]	1400	37	6.2	7.6	29	0.70	0.10	3.1	6.5	6.1	2.1
Treated-6 [Var River (Nice)]	1800	33	6.6	8.5	38	0.73	0.11	3.0	2.8	6.1	2.3

Radionuclide and Trace Element Concentrations in Northeast Pacific Ocean Waters and Zooplankton

W. B. Silker and D. E. Robertson

In a continuing effort to characterize fallout distribution in the North Pacific Ocean and to use these fallout radionuclides as tracers of vertical transport, two sampling cruises were conducted, one during April 1970 in the Gulf of Alaska and Bering Sea and the other off the Washington coast in July. During the April cruise zooplankton and seawater samples were also collected for radionuclide and trace element analysis.

The temperature structure of the water column in the Gulf of Alaska was typical of spring conditions in this area, with cooler surface temperatures gradually increasing with depth a few tenths of a degree Celsius at 200 meters. This was reflected by relatively uniform concentrations of ^7Be , $^{95}\text{ZrNb}$, ^{106}Ru and ^{144}Ce in samples obtained to depths of 40 meters. This rapid vertical transport would be expected in the absence of a thermal gradient within the water column. Surface water concentrations of ^7Be and $^{95}\text{ZrNb}$ collected from deep water stations in the latitude belt between 52° and 54° N and between 142° and 166° W were within $\pm 15\%$ of being the same, which lends credence to our previous hypothesis that average fallout deposition is uniform across a given latitude at sea. Samples taken from shallow water above the continental shelf ($54^\circ 49'$ N, $166^\circ 05'$ W) contained less ^7Be , but equal amounts of fission products,

compared with samples taken on an adjacent station in deep water.

The sampling areas at about 47° N latitude off the Washington Coast during July were occupied during a period of intrusion of a warm surface water mass, possibly an extension of the Kuroshio current, nearly to the coastline. The thickness of the warm water layer (15.0 to 15.9°C) varied from greater than 30 meters at 128° W to less than 20 meters at 125° W, and was characterized by a uniform concentration of ^7Be . The concentrations of the fission products $^{95}\text{ZrNb}$, ^{106}Ru and ^{144}Ce , however, were consistently highest at the surface, and decreased with depth, as though there had been a recent deposition of large quantities of fission products. Further evidence of such an occurrence is derived from the fact that surface concentrations of ^7Be were nearly the same as in Alaskan waters, yet the fission product concentrations were three to four times higher off the Washington coast. Vertical diffusion coefficients calculated from changes in radionuclide concentrations with depth for the four profiles averaged $7\text{ cm}^2/\text{sec}$ for ^7Be and 1 to $2\text{ cm}^2/\text{sec}$ for the three fission products. Total inventories of the radionuclides in the four water columns sampled off the Washington coast were very uniform, and averaged $17,000$, $25,000$, $17,600$ and $49,000\text{ dpm/m}^2$ for ^7Be , $^{95}\text{ZrNb}$, ^{106}Ru and ^{144}Ce , respectively.

Radionuclide concentrations in 500 grams of zooplankton collected by standard surface trawling were below the limits of detection of our sensitive multidimensional gamma-ray

spectrometers. The present radio-nuclide concentrations in the North-east Pacific Ocean and in zooplankton are extremely low and present no hazard to marine organisms or to human populations consuming marine organisms.

Trace element concentrations in the Northeast Pacific Ocean and Bering Sea

were typical of open Pacific Ocean waters. The concentrations of the conservative elements Cs, U and Sb were slightly lower than oceanic waters, but can be explained by the lower salinities in the sampling areas.

Radionuclide Distributions in the North Pacific Ocean

Date	Position		Sampling Depth, meters	Temp., °C	dpm/m ³			
	N	W			⁷ Be	⁹⁵ ZrNb	¹⁰⁶ Ru	¹⁴⁴ Ce
Gulf of Alaska and Bering Sea								
4/7/70	52°44'	141°57'	0	6.0	261±13	129±1	96±8	334±84
4/8/70	53°37'	149°06'	0	4.9	224±10	110±1	71±6	224±72
4/10/70	54°46'	155°03'	0	4.6	265±15	137±1	105±9	491±99
4/11/70	54°00'	161°54'	0	5.2	239±9	149±1	98±6	149±62
			11	5.3	170±10	110±1	83±8	128±62
			23	5.3	244±12	162±1	130±8	112±77
4/12/70	54°49'	166°05'	0	3.5	131±13	125±1	107±8	96±81
			20	3.6	138±10	96±1	81±6	62±45
			39	3.7	191±12	124±1	113±8	112±76
4/12/70	54°18'	166°21'	0	4.3	233±13	127±2	91±9	131±86
			18	4.2	152±8	90±1	61±5	98±48
			37	4.5	218±9	112±1	77±6	168±69
Washington Coastal Waters								
7/25/70	46°50'	128°03'	0	15.2	248±8	490±3	309±10	656±75
			12	15.1	260±8	383±1	229±6	733±59
			27	15.0	256±6	364±1	223±6	573±47
			48	13.1	118±10	253±1	148±6	489±57
			72	11.1	109±8	102±1	99±5	242±59
			97	11.1	69±4	64±1	73±4	289±67
7/26/70	46°59'	127°42'	0	15.4	244±10	517±2	286±9	695±87
			22	15.2	265±7	459±1	256±7	574±59
			39	14.1	175±7	339±1	175±8	449±60
			60	12.8	142±10	121±1	98±8	183±72
			84	12.1	Sample Lost			
7/26/70	46°50'	126°53'	0	15.2	254±15	533±2	324±10	730±93
			29	14.2	170±10	310±1	138±3	577±43
			48	11.3	111±9	143±1	96±5	312±62
			74	10.2	72±7	49±1	48±3	197±30
			98	10.1	94±8	41±1	69±6	188±50
7/27/70	46°51'	125°52'	0	15.9	312±43	619±6	481±16	675±120
			22	14.5	193±8	395±1	186±7	581±52
			39	11.5	121±12	173±1	113±7	384±76
			61	11.2	114±7	70±1	77±6	206±46
			84	10.2	65±5	36±1	66±6	322±81

Radionuclide Concentration in Northeast Pacific Ocean
Zooplankton - April 1970

<u>Sample</u> <u>Wt/g</u>	<u>Position</u>	<u>Depth</u>	<u>Concentration - dpm/kilogram wet weight</u>							
			<u>⁵⁴Mn</u>	<u>⁶⁰Co</u>	<u>⁶⁵Zn</u>	<u>⁹⁵ZrNb</u>	<u>¹⁰⁶Ru</u>	<u>¹³⁷Cs</u>	<u>¹⁴⁰Ba-La</u>	<u>¹⁴⁴Ce</u>
500	54°49'N 166°05'W	Surface	<20	<1	<20	<70	<15	<20	<180	<1000

Trace Element Concentration in Northeast Pacific Ocean
and Bering Sea Waters - April 1970

<u>Sample</u>	<u>Position</u>	<u>Depth</u>	<u>Concentration - µg/liter</u>						
			<u>Zn</u>	<u>Fe</u>	<u>U</u>	<u>Cs</u>	<u>Sb</u>	<u>Co</u>	<u>Sc</u>
1	52°43.5'N 141°57'W	Surface	10	<2	2.9	0.26	0.15	0.0071	0.0003
2	53°36.9'N 149°06'W	Surface	138*	<1	3.1	0.28	0.17	0.0230	0.0008
3	54°46'N 155°03'W	Surface	18	<3	2.7	0.23	0.13	0.0070	0.0004
4	54°00'N 161°54'W	Avg. Top 50 meters	30	25	2.8	0.24	0.20	0.029	0.0018
5	54°49'N 166°05'W	Avg. Top 50 meters	12	8	2.8	0.24	0.19	0.019	0.0010
6	54°18'N 166°21'W	Avg. Top 50 meters	9	<12	2.9	0.26	0.17	0.027	0.0008

* Possible contamination

GEOCHEMICAL OCEANS SECTIONS STUDY (GEOSECS)

GEOSECS is a multidisciplinary detailed study of the properties and constituents of the same water samples to be taken from depth profiles on tracks along the western boundaries of the major oceans. Battelle-Northwest's contribution includes programs to measure trace element, ^7Be , and fallout radionuclide distributions in order to determine their world-wide oceanic deposition and calculate the vertical transport of different water masses from the vertical concentration gradients. To date two intercalibration cruises, one in the Pacific and one in the Atlantic, have been conducted for the purpose of coordinating the different GEOSECS programs and intercomparing the methods and precision for measuring the multitude of oceanic properties to be studied. Results of an interlaboratory comparison of trace element methodology are reported, together with radionuclide distributions in water columns sampled in the Atlantic Ocean.

Trace Element Distribution and Behavior in a Pacific Ocean Column:
Interlaboratory Calibration and Evaluation

D. E. Robertson, D. W. Spencer*
and K. K. Turekian**

In September 1969, the first pre-GEOSECS intercalibration cruise was conducted in the Northeast Pacific Ocean off San Diego, California. During this cruise sixteen 30-liter seawater samples were collected between the surface and 4000 meters depth for trace element analysis. Samples were split into four aliquots and treated as follows: (1) unfiltered-unacidified, (2) unfiltered-acidified, (3) filtered-unacidified, and (4) filtered-acidified. In this manner the effects of filtration and

acidification were evaluated. Filters used were 0.5 μ Nuclepore[®] backed by 0.5 μ Millipore[®] filters. Addition of concentrated hydrochloric acid reduced the pH to 1.7. The following important conclusions were derived from this study:

- Filtration did not significantly alter the concentrations of any of the elements, except possibly for a slight decrease in iron concentrations. Suspended particulate material in the water column was very low, only a few tens of micrograms per liter.
- Acidification of seawater during storage was necessary to prevent serious losses of Fe, Zn, and Co by adsorption on the polyethylene container surfaces.
- Strontium, Rb, U, Cs and Sb were all homogeneously distributed throughout the water column and were essentially proportional to the salinity.

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- Zinc, Fe, Ni and Cu exhibited sharp two to threefold increases in concentration near a depth of 700 meters which were correlated with the oxygen minimum layer. The accumulation at this depth probably resulted from the decomposition, oxidation, and dissolution of the continual fine rain of particulate detritus sinking toward the ocean floor. The cobalt concentration showed a well-

defined two or threefold increase between 200 and 500 meters, while the scandium increased gradually by a factor of four between the surface and 4,000 meters. The differences in the rates of concentration increase with depth between the elements may be due to different rates of regeneration of the elements by the decomposition of the detritus.

Interlaboratory Analyses of Trace Element Concentrations
in a Pre-GEOSECS Northeast Pacific Ocean Depth Profile -
September 1969

28°29' N Lat., 121°38' W Long., Water Depth: 4115 m

Element	Laboratory	Avg. Conc. (Acidified) µg/liter	Avg. Conc. (Unacidified) µg/liter	Percent Difference
Sr	Battelle	8240	8240	0
Sr	Woods Hole	8330	8250	0.9
Sr	Yale	8100	8180	-0.9
Zn	Battelle	2.86	1.71	40
Zn	Woods Hole	3.78	1.73	54
Zn	Yale	7.07	5.47	23
U	Battelle	3.49	3.38	3
U	Yale	3.18	3.14	1
Cs	Battelle	0.30	0.31	-2
Cs	Yale	0.34	0.34	0
Co	Battelle	0.030	0.021	30
Co	Yale	0.083	0.106	-28
Fe	Woods Hole	5.57	1.50	73
Fe	Yale	4.97	3.56	28
Sb	Battelle	0.223	0.218	2
Sc	Battelle	0.00058	0.00050	14
Cu	Woods Hole	1.37	1.12	18
Ni	Woods Hole	2.29	2.39	-4
Rb	Yale	124	123	0.8

- Good laboratory agreement was achieved for Sr, U, and Cs. However, the Co, Zn and Fe results of Yale are high, probably because the samples were contaminated during analysis. The Co concentrations measured by Battelle-Northwest are typical of previous oceanic values determined by Robertson, but are approximately 25 times lower than earlier measurements by Turekian in Central Pacific Ocean waters.

Because of the discrepancies in the trace element work and in measurements of other constituents by various investigators participating in the Pacific cruise, another intercalibration cruise was conducted in August-September 1970 in the Northwest Atlantic Ocean to perfect sampling and analytical techniques. Seawater samples collected during this cruise are presently being analyzed after the manner of the Pacific Ocean samples.

Radionuclide Distributions in Northwest Atlantic Ocean Water Columns

W. B. Silker

During the second pre-GEOSECS cruise aboard the R/V Knorr at an Atlantic Ocean station at 36° N, 68° W, major effort was expended on the measurements of radionuclides contained in the water column. Three vertical profiles, to depths of about 100 meters were sampled during station occupation with equipment used extensively in our other oceano-

graphic work. Equipment modification for GEOSECS included (1) simultaneous sampling from all depths using a bundled sampling hose arrangement and five Battelle Large Volume Water Samplers, thus significantly reducing station time required for sampling; and (2) a pneumatic depth measuring system for real time evaluation of sampling position.

The inventories of four radionuclides contained by the three water columns show internal consistency, but are far different from those observed in Pacific Ocean water. Atlantic Ocean columns contained 2 to 2.5 times more ^7Be and $^{95}\text{ZrNb}$ and about 1.5 times more long-lived ^{106}Ru and ^{144}Ce than Pacific samples collected at slightly higher latitude.

Vertical eddy diffusion coefficients (K_z) calculated from changes in the concentration of ^7Be and $^{95}\text{ZrNb}$ with depth were essentially the same between the depth interval of 60 to 100 meters, varying between 1.1 to 1.2 cm^2/sec . This location was well within the thermocline. Between 40 and 60 meters, near the top of the thermocline, K_z values were somewhat higher, 1.5 to 1.8 cm^2/sec , presumably because of turbulence generated by winds at the sea surface.

It is proposed that the differences in ratios of radionuclides within the water columns, to be sampled in the expanded GEOSECS tracks, together with measurements of their airborne concentration and deposition rates can form the basis for calculations of three-dimensional water mass transport in the world's oceans.

Radionuclide Inventories in Atlantic Ocean Water
Pre-GEOSECS Station - 36° N, 68° W (dpm/m³)

<u>Sampling Date</u>	<u>⁷Be</u>	<u>⁹⁵ZrNb</u>	<u>¹⁰⁶Ru</u>	<u>¹⁴⁴Ce</u>
8/27/70	47,500	48,500	25,000	77,100
8/28/70	40,600	38,800	18,500	62,700
8/29/70	40,000	48,200	25,300	71,900
Average	42,700	45,200	22,900	70,600

Vertical Radionuclide Concentration Gradients in Atlantic
Ocean Water Column (8/27/1970 at 36° N, 68° W)

IN-CLOUD SCAVENGING

Rainwater concentrations of the cosmogenic radionuclides ^{24}Na , ^{38}Cl and ^{39}Cl and their production rates from argon were measured in studies of the rates and mechanisms of in-cloud processes such as raindrop nucleation and precipitation scavenging.

Studies of In-Cloud Scavenging Rates and Mechanisms Using Short-Lived Cosmogenic Radionuclides

J. A. Young, R. W. Perkins,
C. W. Thomas, N. A. Wogman and
B. C. Scott*

The production rates of the cosmogenic radionuclides are known to increase by 2 to 3 orders of magnitude between ground level and near the top of the atmosphere, and it appears that the atom production rates of ^{24}Na : ^{38}Cl : ^{39}Cl are about 1:20:30 at precipitation formation altitudes. Assuming that these production ratios are correct, it can be shown that the ^{39}Cl : ^{38}Cl disintegration rate ratio would be restricted to a range of 1:1 to 1.5:1 and the ^{24}Na : ^{38}Cl ratios would be restricted between 0.0021:1 and 0.05:1 for a steady state production with no vertical movement of air but with precipitation scavenging allowed. Thus, the ratio of the relatively long-lived ^{24}Na to the short-lived ^{38}Cl in rainwater provides a means of determining the time period between any previous atmospheric cleansing, due to precipitation scavenging, and the present in-

cloud nucleation while the ^{39}Cl : ^{38}Cl ratio can be utilized to determine the raindrop development time.

The short-lived cosmogenic radionuclides ^{24}Na , ^{38}Cl and ^{39}Cl were separated from 61 rainwater samples collected during the winter and spring of 1969-1970 at Richland and Quillayute, Washington, by passing the water through a fiberglass filter to remove particulates, an Al_2O_3 bed to remove ^{214}Pb and ^{214}Bi , a cation bed to remove ^{24}Na , and an anion bed to remove ^{38}Cl and ^{39}Cl . The filter and the resin beds were analyzed immediately using multidimensional gamma-ray spectrometers.

The cosmogenic radionuclide activities in the rainwater samples showed an inverse relationship between precipitation rate and activity. This indicates that at high rainfall rates the raindrops grew significantly more by condensation, which would dilute the radionuclides already present in the droplet, than in the case of light rains. In light rains growth by coalescence may be relatively more important. There is also an inverse relationship between the ratio of the long-lived to the short-lived cosmogenic radionuclides and the precipitation rate. The ^{39}Cl / ^{38}Cl and ^{24}Na / ^{38}Cl ratios are higher

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at lower precipitation rates. After incorporation in a raindrop, the cosmogenic radionuclides decay away, with the longer-lived radionuclide decaying slower, so the ratio of the long-lived to the short-lived radionuclide increases with time. Therefore, the higher $^{24}\text{Na}/^{38}\text{Cl}$ and $^{39}\text{Cl}/^{38}\text{Cl}$ ratios in light rains indicate a longer time between drop formation and arrival at ground level in light rains than in heavy ones. If it is assumed that essentially all the radionuclides present in a raindrop are incorporated at the time of the original nucleation of the droplet around an aerosol particle, it can be calculated that times of 20 to 120 minutes are required to reach ground level. If significant amounts of cosmogenic radionuclides are incorporated after nucleation, then the

times to reach ground level would be longer than calculated.

The heights and bases of the rain clouds were estimated using ceilometer and radiosonde data, therefore the total cosmogenic radionuclide content in the cloud could be calculated using the measured atmospheric concentrations. From the radionuclide concentrations in clouds and rain arriving at ground level, rainout coefficients of 10^{-4} sec^{-1} to $5 \times 10^{-3} \text{ sec}^{-1}$ were calculated, where the rainout coefficient is defined as the fraction of the radionuclides scavenged per unit time. These high rainout coefficients indicate that scavenging is very rapid, with the radionuclide bearing aerosol being scavenged within a few minutes at the location of raindrop nucleation.

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^{38}Cl Deposition Rates, Decay Corrected
to Nucleation Times and Rainout Coefficients

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Time Period Between Previous Atmospheric
Cleansing and Present In-Cloud NucleationThe High Energy Proton Spallation
of Argon

N. A. Wogman and R. L. Brodzinski

The high-energy nucleon spallation of argon in the earth's atmosphere produces cosmogenic radionuclides such as ^7Be , ^{18}F , ^{22}Na , ^{24}Na , ^{27}Mg , ^{28}Mg , ^{28}Al , ^{34}mCl , ^{37}S , ^{38}Cl and ^{39}Cl . These radionuclides can serve as tracers of precipitation mechanisms and short-term atmospheric mixing processes. However, this requires a knowledge of the radionuclide production rates in the atmosphere as a function of altitude. To determine the expected production ratios of these potential atmospheric tracers, their atmospheric production from argon was simulated with a cyclotron

bombardment experiment. The production cross sections for the formation of several of these radionuclides by proton spallation of argon at 310, 425, and 578 MeV were measured. These measured values were then used as normalization points for excitation functions (cross section versus energy curves) which had been calculated by nuclear spallation systematics. Excitation functions were thus generated for the radioisotopes ^{22}Na , ^{24}Na , ^{27}Mg , ^{37}S , ^{38}Cl and ^{39}Cl .

From earlier measurements of argon spallation products formed in high pressure argon cylinders which were flown at 19.8 km, it was estimated that between 3.3 km and sea level at 46°N latitude the production rates of ^{24}Na : ^{38}Cl : ^{39}Cl are in the ratio of

1:20:30. Assuming that our calculated shapes of the excitation functions for formation of these radioisotopes from argon are fairly accurate, their production ratios with 250 MeV protons would be 1:22:28. Similar results would be obtained for high energy neutron reactions. If the average effective nucleon energy is substantially greater than 250 MeV a contribution from muon reactions is necessary to achieve a ratio with sufficient ^{38}Cl and ^{39}Cl to equal 1:20:30. The contribution of the muon component to the spallation yield of atmospheric argon is being evaluated.

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RADIOANALYTICAL PROCEDURE DEVELOPMENT

Radiochemical separation procedures which are compatible with and augment advanced instrumental analysis technology are required to provide a complete quantitative analysis of a radionuclide mixture. Although some of the developed methods are designed to separate a specific radionuclide, the major effort is directed toward group separation procedures.

Analytical Procedure for Cosmogenic Radionuclides in Rainwater

C. W. Thomas, R. W. Perkins and
J. A. Young

The short-lived cosmogenic radionuclides ^{38}Cl (37.3 min), ^{39}Cl (55 min), and ^{24}Na (15 hr) are particularly attractive as tracers of precipitation scavenging processes because of their half-lives, their attachment to natural aerosols, and their unique decay schemes which allow detection by multidimensional gamma-ray spectrometry. The major difficulty in using these cosmic-ray-produced radionuclides as tracers is their very low concentration in precipitation and the relatively high concentration of associated radon daughter products.

In the initial procedure, large volumes of rainwater were collected and cosmogenic chlorine isotopes were removed by AgCl precipitation. This procedure was not conducive to short sampling periods and was replaced by a continuous separation method using ion exchange resins. Although the latter technique allowed short sampling times, interference problems still re-

mained from ^{214}Bi and ^{214}Pb and sporadic channeling of the thin resin beds. Laboratory and field evaluations during the 1970 precipitation sampling period indicated that chromatographic grade Al_2O_3 would remove greater than 80% of the ^{214}Pb and ^{214}Bi in the precipitation with no measurable removal of ^{38}Cl , ^{39}Cl and ^{24}Na . Additional tests also showed that by using 200 to 400 mesh resin a uniform bed could be reproduced with excellent retention efficiencies and without large curtailment of flow rates. By combining an Al_2O_3 scavenging bed with the resin technique,, a procedure has been developed for the continuous separation of ^{38}Cl , ^{39}Cl , and ^{24}Na from the radon daughters, ^{214}Pb , and ^{214}Bi , and most fission products in large-volume rain samples.

In field studies the filters, Al_2O_3 and resin beds are held by a recently designed plastic polyvinylchloride (PVC) rainwater sampler. The rainwater is filtered through a particulate filter assembly, a 1 cm thick by 20 cm diameter bed of Al_2O_3 (Woehm Neutral, Aluphor Chemicals), a 0.64 cm thick by 28 cm diameter bed of Dowex 50 $\times$ 8 (H^+ form, 200 to

400 mesh), and a 0.64 cm thick by 28 cm diameter bed of Dowex 1 x 8 (Cl^- form, 200 to 400 mesh). The particulate filter assembly which consists of a glass fiber filter (AP-2030500, Millipore) and a sharkskin filter (Schleicher and Schuell) is

also used to back the Al_2O_3 and resin beds. Flow volumes of 20 to 30 liters/minute can be processed providing sample volumes from which 10% counting statistics can be obtained in periods of less than 1 hour for ^{38}Cl , ^{39}Cl , and ^{24}Na .

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Large-Volume Rainwater Sampler for the Continuous
Removal of ^{38}Cl , ^{39}Cl , and ^{24}Na from ^{214}Bi , ^{214}Pb ,
and Fission Products

The Separation of Plutonium and
Americium from Ashed Bone Samples

P. O. Jackson

The measurement of ^{241}Am in bone specimens is difficult due to the calcium phosphate matrix which causes serious interferences with standard separation techniques. For studies involving the distribution of plutonium and americium in organs of animals exposed to airborne concentrations of these radionuclides, a measurement of the Pu/Am ratio and the total Pu content will yield the americium concentration.

A technique has been developed for the separation of Pu and Am from ashed bone samples based on coprecipitation of the actinides on ferric hydroxide at a specific pH. Other techniques based on cation or anion exchange and solvent extraction with phosphate esters or high molecular weight amines from several chelating agents all proved ineffective due to low recovery or poor separation from inert material. In the present technique iron hydroxide-phosphate gel which forms at a pH of 3.4 to 3.5 is used to quantitatively carry Am^{+3} and Pu^{+4} . Calcium phosphate does not tend to precipitate until a pH of 4.2. Three repetitive precipitations give a final ferric hydroxide precipitate that is free of calcium phosphate when up to 2 grams of bone ash in solution were analyzed.

Iron is subsequently removed by cation exchange. After the Fe is loaded on the cation columns from 1M HCl, it is eluted with 3M HCl and the actinides are stripped using

7M HCl. The specimen is then mounted directly on a watch glass for alpha energy analysis. A uniform deposit (<0.3 mg) is formed by adding 50 μg of zirconium carrier when mounting the final specimen. This uniform deposit provides good alpha energy resolution and a total alpha assay. The technique of adding about 10% Zr by weight to the deposit also gives much more reproducible and higher counting efficiencies.

Optimum procedural conditions were determined using ^{239}Pu and ^{241}Am tracers or high specific activity samples added to blank bone solutions. Recoveries for actual bone samples with ash weights ranging from 10 mg to 3.3 g averaged 100%. A comparison of the alpha spectral analyses of sources prepared directly from ^{241}Am and ^{239}Pu and from tracer solutions run through the procedure gave Pu:Am ratios which agreed within $\pm 10\%$. The method should also be useful for other trivalent actinide separations from ashed fecal material.

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RADIOACTIVE FALLOUT RATES AND MECHANISMS

The concentrations of trace elements, cosmogenic radionuclides, and nuclear-weapons-produced radionuclides were measured in the air from ground level to 19 km and in rain and seawater in order to determine the origin and chemistry of the atmospheric aerosol and to study the rates of atmospheric and oceanic mixing, the rates of air-sea interchange, and the rates and mechanisms of precipitation scavenging.

Atmospheric Fallout

C. W. Thomas

The atmospheric concentrations of 20 radionuclides in ground level air were measured continuously during 1970 at Point Barrow, Alaska (71° N), Richland, Washington (46° N), Barbados British West Indies (13° N), and Rio de Janeiro, Brazil (23° S). The concentrations of the nuclear-weapons-produced radionuclides in the northern hemisphere have increased steadily from 1967 through 1970, primarily because of the high yield thermonuclear tests conducted by the Chinese at Lop Nor (45° N) on 6/17/67, 12/24/67, 12/27/68, 9/29/69 and 10/15/69. During 1970 more than 95% of the nuclear-weapons-produced radionuclides present in northern hemispheric air originated from the Chinese tests. The radionuclide concentrations at Rio de Janeiro (23° S) increased sharply after the French tests conducted in the South Pacific in May 1970. The radionuclide concentrations reached values comparable to those measured in 1968 after the 1968

French test series which included two thermonuclear detonations with an accumulated yield of four megatons. In contrast to the 1968 tests, large amounts of ^{88}Y were produced by the May 1970 French tests.

Air samples collected by RB-57F aircraft at altitudes from 12.2 to 18.3 km over Spokane, Washington (48° N) contained large amounts of ^{140}Ba - ^{140}La on 10/18/69 and on 9/28/70, 9/29/70, 9/30/70 and 10/1/70, but not on 4/2/70. The ^{140}Ba - ^{140}La present in the 10/18/69 samples came from the Chinese thermonuclear test of 9/29/69. By 4/2/70 the ^{140}Ba - ^{140}La had decayed away, but there were still large amounts of slightly longer-lived radionuclides such as ^{95}Zr - ^{95}Nb and ^{103}Ru present. The ^{140}Ba - ^{140}La in the samples collected from 9/28/70 through 10/1/70 probably came from the French thermonuclear tests conducted in the southern hemisphere in late May of 1970. Ground level air samples collected at Barbados (13° N) and Rio de Janeiro (23° S) in June 1970 contained ^{140}Ba - ^{140}La .

Atmospheric Stable Elements as
Tracers of Precipitation Scavenging

L. A. Rancitelli, R. W. Perkins,
T. M. Tanner and C. W. Thomas

Material, both organic and inorganic, is injected into the atmosphere from a variety of natural and man-made sources. These trace constituents, which are often characterized by very specific chemical compositions, mix with the various air masses and are eventually removed by dry deposition or precipitation scavenging. During their residence time in the atmosphere, they are potential tracers of the atmospheric processes responsible for their transport and deposition, as well as potential indicators of the contribution of each aerosol source to the total aerosol inventory. However, the use of these aerosol particles as tracers of atmospheric processes has been rather limited due to the lack of practical analytical techniques capable of measurements at the ultra-low levels frequently encountered. An instrumental technique which allows direct multi-element analyses following neutron activation has recently been developed and employed in defining the origin, the average concentrations, and the concentration gradients of several elements in the atmosphere. An extension of this effort has been the development and application of similarly sensitive techniques for rainwater analyses which also provide information on the aerosol origins in precipitation, and on the temporal variations in elemental concentra-

tions. The use of this information on trace element concentrations in the air, together with measurements of the trace elements in precipitation, provides a basis for calculating the atmospheric cleansing rate due to precipitation scavenging. It also provides much of the information and technology necessary for employing trace elements, which are intentionally added to the atmosphere, as specific tracers of the atmospheric processes.

The concentrations of 17 elements were determined in samples of rainwater collected at Quillayute, Washington. A comparison of concentrations of these elements normalized to the Fe content of the earth's crust indicated seven elements, Ag, As, Br, Cl, Sb, Se and Zn, were significantly enriched in rainwater relative to the earth's crust. The rainwater concentrations of five elements, Co, Fe, Sb, Sc and Zn, were used in combination with average trace element concentrations in the atmosphere and meteorological data obtained during the rainstorm to estimate scavenging coefficients. The three elements, Co, Fe and Sc, which evidently originate from the earth's crust yielded rather constant scavenging coefficients of 5 to $10 \times 10^{-5} \text{ sec}^{-1}$. Antimony and zinc yielded higher values of $30 \times 10^{-5} \text{ sec}^{-1}$ and $160 \times 10^{-5} \text{ sec}^{-1}$, respectively, possibly reflecting the fact that the concentrations of these elements, which evidently originate from anthropogenic sources, are highly variable in the atmosphere.

Comparison of Elemental Abundances in Rainwater and the Earth's Crust
(Normalized to Fe = 50,000 ppm)

<u>Element</u>	<u>Earth's*</u> <u>Crust</u>	<u>Rainwater</u>	<u>Rain/Earth</u>
Ag	0.07	1400	2×10^4
Cl	130	1,400,000	1.1×10^4
Se	0.05	100	2×10^3
Br	2.5	3900	1.6×10^3
Sb	0.2	230	1.2×10^3
As	1.8	1900	1.1×10^3
Zn	70	28,600	4.1×10^2
Cu	55	4900	89
Cs	3	100	33
Rb	90	<2900	<32
Na	28,300	810,000	29
Cr	100	<2900	<29
K	25,900	470,000	18
Mn	950	3700	3.9
Co	25	64	2.6
Sc	22	4	0.2

* *Mason, B. (1966)*

Inorganic Emissions from the Hanford
Steam Plant

L. A. Rancitelli

It is generally recognized that the oceans and crustal surfaces serve as the major sources of the atmospheric aerosol. However, when one considers the atmospheric inventory of specific trace constituents, deviations from natural abundances are

evident which can be attributed to man's activity. While a prime example is lead which arises as an oxidation product in automobile exhaust, other elements in the atmosphere can also be attributed to anthropogenic origin. The burning of fossil fuels must certainly be considered as one of the major sources of stable tracer elements in the atmosphere. Although their contribution to the worldwide

inventory of a specific element may not be great, they can cause significant changes on a local level.

An effort has been made to establish the inorganic emission of a Hanford steam plant by characterizing the trace element content of coal and ash. The absolute concentrations or upper limits to the concentrations of 12 elements, As, Br, Co, Cr, Fe, Hg, K, Na, Rb, Sb, Sc, and Se, were determined in samples of the coal and ash by instrumental neutron activation analysis. A material balance of these elements observed in the coal and ash reveals that the transition metals, Cr and Fe, and the alkali metals, Na and Rb, appear to be conserved while Hg, Sb, Se and Sc and to a lesser degree Co are depleted in the ash relative to the coal. Based upon the 20,000 tons/year consumption rate and the dry ash production of 1200 tons/year of this plant we can estimate that approximately 2.9 pounds of Hg, 31 pounds of Se and 2.6 pounds of As are emitted by the plant per year. While these represent rather nominal amounts of these materials, an extrapolation of these values to the annual consumption rate of coal in the United States indicates that about 30 tons of As and Hg and over 300 tons of Se per year will be placed in the

atmosphere by the burning of coal. These estimates may be conservative since the coal employed in the Hanford steam plant is a higher grade than that normally employed for power production, and hence might reasonably be expected to contain smaller amounts of Se and Hg along with its lower sulfur content.

Elemental Content of Coal and Ash			
(Parts per million)			
<u>Element</u>	<u>Coal</u>	<u>Ash</u>	<u>Ash/Coal</u>
As	<0.05	<0.05	---
Br	<0.5	---	---
Co	0.74	8.1	11
Cr	1.78	32.0	18
Fe	2.16	38.0	18
Hg	0.072	<0.01	<0.14
K	<970	<250	---
Na	620	10,000	16
Rb	1.38	29.8	22
Sb	0.10	0.74	7.1
Sc	0.85	4.6	5.4
Se	0.97	3.2	3.3

Cosmogenic Radionuclide Production
Rates in Argon in the Atmosphere
from 3 to 19 km

J. A. Young, C. W. Thomas and
N. A. Wogman

For the past three years air filter samples have been collected by aircraft for the determination of atmospheric radionuclide concentrations. Among the radionuclides measured have been the cosmogenic radionuclides ^{24}Na (15 hr), ^{38}Cl (37 min), and ^{39}Cl (55 min). These cosmogenic radionuclides are produced at a relatively constant rate by spallation reactions of cosmic-rays with atmospheric argon.

In order to determine whether the measured cosmogenic radionuclide disintegration rates corresponded to the actual production rates, metal spheres containing 17 to 42 standard cubic meters (SCM) of argon under pressure were flown at altitudes ranging from 3 to 19 km. Since the radioactive spallation product atoms produced in the argon are recoiled or electrostatically transferred to the wall, the inner surface of each sphere was lined with a meteorological balloon. At the end of each flight the argon was vented through a charcoal-impregnated filter. The

filters and rubber liners were then composited and the cosmogenic radionuclide concentrations measured by direct analysis on a multidimensional gamma-ray spectrometer that utilizes two 34 cm diameter by 15 cm thick NaI (Tl) crystals. In addition to ^{24}Na , ^{38}Cl and ^{39}Cl , the cosmogenic radionuclides ^{18}F , ^{28}Mg , ^{34}mCl , and ^{38}S were measured in argon in some of the flights at 18 and 19 km.

The measured ^{24}Na production rates are almost identical to the measured atmospheric disintegration rates, indicating that the air filters are removing essentially all of the ^{24}Na from the air. The ^{38}Cl production rates, however, are much higher than the disintegration rates. This indicates either that the short-lived ^{38}Cl and ^{39}Cl radionuclides do not have time to become completely attached to the atmospheric aerosol, and are not being completely removed from the air by the filters, or that the production rate is enhanced in the argon spheres due to low energy radiation produced by reactions of cosmic rays with the aircraft and the metal spheres. Higher energies are required for ^{24}Na production, so the ^{24}Na production rate would not necessarily be enhanced by this latter effect.

Cosmogenic Radionuclide Production Rates in Argon

DATE	LATITUDE (°N)	LONGITUDE (°W)	ALTITUDE (Km)	PRODUCTION RATE IN ATOMS/MIN/SCM ARGON								DISINTEGRATION RATE IN AIR DPM/1.08X10 ² SCM AIR			
				¹⁸ F	²⁴ Na	²⁸ Mg	^{34m} Cl	³⁸ S	³⁸ Cl	³⁹ Cl	²⁴ Na ³⁸ Cl	³⁹ Cl ³⁸ Cl	²⁴ Na	³⁸ Cl	
5-16-69	35-47	107-117	19.8		6.6 ± 1.4		12.4 ± 1.4		116 ± 17	155 ± 9	0.040 ± 0.010	1.34 ± 0.21			
8-28-69	35-47	107-117	18.3	1.4 ± 0.5	7.7 ± 0.4	2.0 ± 0.3	13.3 ± 1.5	5.0 ± 1.0	121 ± 9	183 ± 5	0.064 ± 0.006	1.51 ± 0.17			
10-28-69	35-47	107-117	18.3		5.4 ± 0.4				105 ± 5	168 ± 10	0.051 ± 0.004	1.60 ± 0.12			
4-21-70	36-47	107-117	18.3	3.0 ± 1.2	7.0 ± 0.5	2.0 ± 1.2	13.1 ± 1.1	3.5 ± 2.3	105 ± 15	178 ± 6	0.067 ± 0.011	1.70 ± 0.30			
7-27-70	46	125	12.2		2.4 ± 0.3				45.5 ± 3.9	77.5 ± 3.2	0.053 ± 0.008	1.70 ± 0.16	3.0 ± 0.1	20.4 ± 1.3	
7-28-70	46	125	9.1		1.4 ± 0.2				31.2 ± 3.1	40.3 ± 3.2	0.045 ± 0.008	1.29 ± 0.16	1.07 ± 0.04	9.1 ± 0.9	
9-28-70	35-46	107-125	9.1		1.6 ± 0.3				35.1 ± 3.8	74.8 ± 3.0	0.046 ± 0.010	2.13 ± 0.24	0.59 ± 0.04	4.0 ± 0.8	
7-29-70	46	125	6.1		0.35 ± 0.05				8.6 ± 1.7	16.1 ± 2.5	0.041 ± 0.010	1.87 ± 0.47	0.35 ± 0.02	8.2 ± 0.6	
10-1-70	46	125	6.1		0.67 ± 0.30				12.5 ± 3.0	13.1 ± 3.1	0.054 ± 0.027	1.05 ± 0.35	0.21 ± 0.02	7.5 ± 0.8	
10-20-70	46	125	6.1		0.57 ± 0.20				6.2 ± 2.0	23.0 ± 2.0	0.092 ± 0.061	3.7 ± 1.2	0.41 ± 0.03	1.9 ± 0.4	
9-30-70	46	125	4.6		0.064 ± 0.030				3.4 ± 1.1	10.4 ± 1.1	0.019 ± 0.011	3.1 ± 1.1	0.10 ± 0.02	0.64 ± 0.32	
10-21-70	46	125	4.6		0.07 ± 0.03				2.6 ± 1.3	10.0 ± 1.0	0.027 ± 0.018	3.9 ± 2.0	0.23 ± 0.02	3.0 ± 0.4	
7-30-70	46	125	3.1		0.08 ± 0.04				1.7 ± 1.1	1.7 ± 1.9	0.047 ± 0.038	1.0 ± 1.3	0.072 ± 0.011	3.7 ± 0.5	
9-29-70	46	125	3.1		0.04 ± 0.03				2.3 ± 1.1	6.9 ± 0.8	0.017 ± 0.015	3.0 ± 1.5	0.040 ± 0.011	0.7 ± 0.3	

Neg 710081-1

Neg 710080-2

Production Rates of ^{24}Na , ^{38}Cl , and ^{39}Cl in Air

Cosmogenic Radionuclide Production Rates in the Atmosphere

J. A. Young, C. W. Thomas,
N. A. Wogman and R. W. Perkins

Spallation reactions of cosmic rays with atmospheric argon produce ^{24}Na (15 hr), ^{38}Cl (37 min), and ^{39}Cl (55 min) at a relatively constant rate. Because of their short half-lives, these radionuclides have proved to be very useful tracers of precipitation scavenging mechanisms. However, before they could be used most effectively, it was necessary to know their atmospheric concentrations. Therefore, in March 1967 we began measuring their activities on air filter samples collected by RB-57F aircraft on IPC-1478 filter paper. The activities were determined using NaI(Tl) multidimensional gamma-ray spectrometers.

The measured atmospheric activities of ^{24}Na , ^{38}Cl and ^{39}Cl increased approximately exponentially with altitude by a factor of 2,000 between ground level and 19 km at 46°N . The ratios of the activities did not change significantly with altitude. The ^{24}Na activity at 18.3 km, 46°N , 117°W , has decreased by about 25% since March 1967. The decrease was presumably caused by the increase in sunspot activity which has occurred since 1967. The flux of cosmic rays of galactic origin is decreased by sunspot activity. The effect is greatest for cosmic rays of low rigidity, so the effect should be greatest at high altitudes. No systematic variation in ^{24}Na activity with time was observed at altitudes below

18.3 km, although the scatter of the experimental points at lower altitudes would have made a time variation difficult to detect.

The measured ^{24}Na disintegration rate at 18.3 km increased by a factor of about 4 between 0° and 60°N . The rate of increase appeared to be significantly lower from 40° to 60°N than from 0° to 40°N . The change in activity with latitude at 15.2 km appears to parallel that at 18.3 km, although there may be a significant difference in the shape of the variation north of 40°N .

Project BOMEX Studies of Atmospheric and Oceanic Mixing and Air-Sea Interchange Using Radioactive Tracers

J. A. Young and W. B. Silker

The activities of cosmogenic ^7Be and several bomb-produced radionuclides were measured in air, seawater, and rain samples during the Barbados Oceanographic and Meteorological Experiment (BOMEX). Because of its relatively short 53 day half-life, ^7Be was confined primarily to the top 50 meters of the sea, so its inventory in the sea could be determined from samples collected between the surface and 100 meters. The inventories of ^7Be and the other radionuclides in the sea varied by a factor of about 2.5 during BOMEX, with most of the variation occurring in the top 30 meters. The observed changes were not due to rain, since the addition of ^7Be by rainfall during the experimental period was only

10 to 15% of the ^7Be decay in the sea. The rapid variations in concentration were probably caused by horizontal transport of water masses of different activities through the BOMEX array.

During the second two weeks of May large amounts of fresh water, presumably Amazon River water, entered the southeast corner of the BOMEX array. Measured surface salinities decreased from around 35‰ at the USCGS Rockaway during the first two weeks of May to as low as 31‰ at the USCGS Discoverer during the last two weeks of June, indicating the presence of around 12% fresh water. The large changes in ^7Be activity could have been caused by intrusions of river waters having high ^7Be activity. Beryllium-7 activities were generally higher when salinities were low. However, the ^7Be activity was not directly proportional to the amount of fresh water present, so the intrusion of fresh water was probably not the only cause of the variation in ^7Be activity. The variations in activity may indicate that ^7Be activities in the ocean are generally not uniform over large horizontal areas, due to differences in the amount of ^7Be transferred across the sea at different locations resulting from different rainfall patterns, air concentrations, and wind speeds.

Air filter samples have been collected on the Island of Barbados since May 1969. These samples indicate that the air concentration of ^7Be varies seasonally by an order of magnitude at this location, with maximum concentrations occurring

around May and June and minimum concentrations occurring in November and December. Therefore, ^7Be inventories in the sea should also change with the time of year. If one assumes that the rate of transfer of ^7Be across the sea surface is proportional only to the air concentration, one can calculate the variation of seawater inventory with time by choosing the proportionality constant so that the calculated inventories agree with the measured inventories. A rate of transfer of 9.52 atoms of $^7\text{Be}/\text{cm}^2$ of sea surface/day/dpm of ^7Be in 10^3 standard cubic meters (SCM) of air gives calculated seawater inventories which agree with measured inventories. These calculations indicate that the maximum and the minimum in the seawater inventory should both occur 2 or 3 months after the maximum and the minimum in the air concentration.

During the BOMEX period the ^7Be inventory in the sea was increasing, on the average, by $0.010 \text{ atom cm}^{-2} \text{ sec}^{-1}$. The rate of addition to the sea was 35% greater than the rate of decay in the sea. Dividing the rate of addition by the average atmospheric concentration gives a value of 0.7 cm sec^{-1} for the deposition velocity. The vertical diffusion coefficients in the surface layers of the ocean can be calculated from the flux across the sea surface and the vertical concentration gradient. The vertical profile measured on May 11, 1969 gives the following values for the vertical diffusion coefficient, K_z .

Depth, meters	K_z , $\text{cm}^2 \text{sec}^{-1}$
0-15	4.3
15-30	4.8
30-50	0.82
75-100	0.60

The activities of ^7Be and the nuclear-weapons-produced radionuclides in air filter samples collected by aircraft during BOMEX did not increase significantly between 0.3 and 9.1 km altitude. On the average there was a minimum in the activity of the radionuclides at 1.5 km, probably because of rainout. Many of the vertical profiles showed a minimum for the activity of the bomb-produced radionuclides but not for ^7Be at some altitude, often 9.1 km. These minimums probably resulted from intrusions of southern hemispheric air, since the activities of the bomb-produced radionuclides were considerably lower in the southern hemisphere at this time, but the ^7Be activity should be about the same in the two hemispheres. Other profiles showed minimums in both ^7Be and the bomb-produced radionuclides at around 9 km and were therefore not due to intrusions of southern hemispheric air. Probably these minimums resulted from recent precipitation scavenging of the air. It is possible that air which was scavenged in the cumulonimbus of the intertropical convergence zone moved northward into the BOMEX array.

Often there were large increases in radionuclide activity with time at some altitude and location. The increases at 12 km during the first 2 weeks of June were particularly large. Back trajectories were calculated for many of the air masses showing high radionuclide activities. It was found that these air masses had originated within the previous few days at latitudes of 30°N or greater. Activities of ^7Be and the bomb-produced radionuclides are generally considerably greater at these higher latitudes.

During the BOMEX experiment, air samples were collected at five elevations from 1 ft to 70 ft above the sea surface aboard Flip, the Rockway and the Discoverer. Many of these profiles showed increasing radionuclide activities with height, due to the removal of the radionuclides at the sea surface. On some of the profiles, however, the increase in activity with height was too small to be measured. When the vertical eddy diffusion coefficients measured by other investigators become available the rate of transfer of ^7Be and other radionuclides across the sea surface as a function of time will be calculated to determine the effect of meteorological parameters such as wind speed upon the flux across the sea surface. The calculated fluxes will also be compared with the average fluxes determined from the measured seawater inventories.

^7Be Inventory in a 1 cm^2 Column
of Seawater

^7Be Disintegration Rate in Air
at Barbados and the Calculated
Seawater ^7Be Inventory

IRON-55 AND LEAD IN THE HUMAN POPULATION OF THE WORLD

This study investigates the transfer of ^{55}Fe from fallout, with special emphasis on the concentrations in man and in food chains leading to human consumption. Transfer is being measured in the general population as a function of age, sex, and diet, including the dilution effect of stable iron. Although ^{55}Fe is being emphasized, the same samples are used to measure Pb , ^{210}Pb , and ^{210}Po in the biosphere.

Iron-55 Concentrations in AlaskanEskimos

C. E. Jenkins

High concentrations of ^{55}Fe in salmon and other oceanic surface feeding organisms have produced high body burdens of this radioelement in populations who eat substantial quantities of marine life. Caribou-eating Eskimos and reindeer-eating Lapps also have substantially higher ^{55}Fe body burdens because of the unique radionuclide concentrating properties of Arctic lichen eaten by the caribou. Concentrations of ^{55}Fe peaked in the following samples in the years indicated: northern salmon-1963, salmon-eating Eskimos and Japanese-1963-64, caribou-1965, caribou-eating Eskimos-1966, Washington cattle-1964, and cattle-eating Washington residents-1966.

Present ^{55}Fe concentrations in Alaskan salmon-eating Eskimos are dropping at a rate slightly less than the salmon. Since salmon are caught and preserved for use throughout the winter months this would be the expected trend. Blood samples taken in

A Comparison of the Relative Decrease of ^{55}Fe in Fallout, Salmon, Rain, and Air (Curves are not corrected for decay.)

March 1969 at Kotzebue, Alaska, show ^{55}Fe concentrations higher than Richland residents but falling rapidly due to the high turnover rate of ^{55}Fe in the oceans. For example, in 1966

the average ^{55}Fe body burdens were 100-fold higher for Kotzebue Eskimos than Richland residents while presently these values are only 10-fold higher.

^{55}Fe in Alaskan Eskimo Blood - March 1969

<u>Location</u>	<u>Sex</u>	<u>^{55}Fe, dpm/g</u>	<u>Fe, mg/g</u>	<u>Specific Activity</u>	<u>Body Burden, nCi</u>
Kotzebue	M	24.8	0.47	52.8	92
	M	15.8	0.56	28.2	59
	M	16.8	0.48	35.0	63
	M	14.9	0.47	31.7	56
	M	12.9	0.53	24.3	48
Kotzebue	F	16.9	0.41	41.2	63
	F	21.2	0.41	51.7	80
	F	5.7	0.51	11.4	21
	F	12.4	0.38	32.6	47
	F	19.5	0.45	43.3	73
	F	1.3	0.37	3.5	5
	F	11.7	0.38	30.8	44
	F	10.0	0.41	24.4	38
	F	27.4	0.47	58.3	103
	F	10.9	0.66	16.5	41
	F	16.2	0.43	37.7	61
	F	13.9	0.42	33.1	52
	F	15.0	0.45	33.3	56
	F	11.7	0.47	24.9	44
	F	25.0	0.69	36.2	94
	F	8.6	0.45	19.1	32
Buckland	M	8.96	0.35	25.6	34
Koyuk	M	36.5	0.60	60.8	137
Noatak	M	23.6	0.54	43.7	89
	M	20.4	0.45	45.3	77
St. Michaels'	M	15.9	0.44	36.1	60
Wales	M	14.0	0.44	31.8	53
Candle	F	21.5	0.44	48.9	80.7
Elim	F	36.4	0.48	75.8	137
	F	20.5	0.46	44.6	77
Noorvik	F	21.3	0.42	50.7	80
Point Hope	F	12	0.46	26.1	45

The Latitudinal Variations of Iron-55 in Man and Cattle

J. C. Langford and C. E. Jenkins

The concentrations of ^{55}Fe and stable iron were measured in a world-wide study of ^{55}Fe radioactivity in human blood to investigate the transfer of ^{55}Fe from fallout through the biosphere, with special emphasis on the concentration levels in man. Iron-55 concentrations in human blood were found to correlate well with the fallout air concentrations as measured by the Health and Safety Laboratory. The exceptions are due to unusual dietary habits or ecology. Iron-55 concentrations in human blood have de-

creased from the 1966 levels (excluding Alaska) to an average of 1.18 dpm/g of blood with a range of 0.25 to 2.60 dpm/g. Alaskan blood averaged 17.4 dpm/g. The concentration of ^{55}Fe was usually lower in women presumably due to the faster turnover of iron in the female. A comparison of ^{55}Fe concentrations in Columbia Basin beef and Columbia River salmon showed that the specific activity of the salmon was 100-fold higher than the beef. The average quantity of salmon eaten yearly by inland populations is sufficient to account for 20% of their dietary intake of ^{55}Fe .

A Comparison of the ^{55}Fe Concentration in Air and Human Blood at Various Latitudes

Particulate Pb, ^{210}Pb , and ^{210}Po
in the Environment

J. C. Langford

Stable lead concentrations have increased in the environment in recent years due to increased industrial output, vehicular densities, and consumption of lead-containing fossil fuels. In addition ^{210}Pb and ^{210}Po are of special interest since both are bone-seeking and are potentially more hazardous to the human body than are some other radioisotopes. Particulate Pb, ^{210}Pb , and ^{210}Po concentrations were measured in a Pacific Ocean depth profile 350 miles west of Newport, Oregon, and in the Atlantic Ocean surface samples collected at various locations between the Bahama Islands and Sierra Leone, Africa.

The particulate ^{210}Po and ^{210}Pb concentrations found in the Pacific Ocean depth profile decreased with depth to 656 meters, below which they remained fairly constant. Radium-226 had the same depth profile except that its concentrations increased from 656 to 1639 meters. Stable lead concentrations decreased with depth to 1639 meters and approached 0.02 μg per liters. Assuming that lead in seawater is primarily from utilization of airborne aerosols from industrial petroleum production, which began a rapid increase in 1940, and

that the lead at 1639 meters is due to lead deposited at the surface in 1940, a good correlation can be made between the concentrations of lead at various depths and the world crude oil production.

In the Atlantic Ocean the average particulate ^{210}Po concentration was 20% higher than the average particulate ^{210}Pb concentration. The particulate stable lead concentration in the Atlantic Ocean decreased ninefold (2.23 ± 0.02 to 0.25 ± 0.03 $\mu\text{g/liter}$) $25^{\circ}38'$ N Lat., $72^{\circ}20'$ W Long. to $24^{\circ}48'$ N Lat., $59^{\circ}20'$ W Long., a distance of 1440 miles. Thereafter, the concentrations slowly decreased approximately 20% to near the African coast. Particulate ^{210}Po and ^{210}Pb concentrations in Atlantic Ocean surface seawater averaged 0.016 and 0.12 dpm/liter, respectively.

It seems that the source of particulate lead in the ocean is primarily of terrestrial origin, presumably by air pollution, since the physical barriers of the Bahama Islands and the Gulf Stream would prevent most eastward flows of mainland rivers from the Florida Coast. Although there are large variations in concentrations between different sources, the urban-industrial concentrations of stable lead in the air and the nearby marine environment are much higher than those from other airborne sources.

Pb, ^{210}Pb and ^{210}Po Concentrations

Atlantic Ocean Water

Location				^{210}Po , dpm/liter	^{210}Pb , dpm/liter	Pb, $\mu\text{g/liter}$	Specific Activity, ^{210}Pb dpm/ $\mu\text{g Pb}$	Distance from U.S.A., statute mi.
N	Lat.	W	Long.					
25°30'		74°		0.007 ± 0.007	0.009 ± 0.002	0.45 ± 0.04	0.020	480
25°38'		72°20'		0.015 ± 0.001	0.016 ± 0.002	2.23 ± 0.02	0.0072	600
25°10'		65°55'		0.014 ± 0.001	0.015 ± 0.002	0.72 ± 0.05	0.021	1,080
24°28'		62°18'		0.018 ± 0.002	0.011 ± 0.002	0.31 ± 0.03	0.036	1,320
24°48'		59°20'		0.017 ± 0.001	0.012 ± 0.002	0.25 ± 0.03	0.048	1,440
8°50'		38°28'		0.022 ± 0.002	0.008 ± 0.002	0.24 ± 0.03	0.030	3,000
5°48'		16°35'		0.017 ± 0.002	0.017 ± 0.002	0.21 ± 0.03	0.081	4,570

Pacific Ocean Water

Depth, meters*	^{210}Po , dpm/liter	^{210}Pb , dpm/liter	Pb, $\mu\text{g/liter}$	Specific Activity, ^{210}Pb dpm/ $\mu\text{g Pb}$
20**	0.14	0.12	2.1	0.057
20	0.049	0.049	0.23	0.21
328	0.031	0.010	0.069	0.14
656	0.014	0.029	0.10	0.29
984	0.024	0.012	0.060	0.20
1639	0.018	0.009	0.036	0.25

* Samples collected at 41° N Latitude and 130° Longitude

** Sample filter coated with some algae

RADIATION INSTRUMENTATION - RADIOLOGICAL CHEMISTRY

These studies are concerned with the development of radiochemical instrumentation and analytical technology, their utilization in radioisotope studies of environmental samples, their application to neutron activation analysis to define stable element versus radioelement behavior, and their use in the elucidation of the routes described by natural, tracer, or waste radioisotopes in the environs.

The Development and Application of a
Beta-Gamma-Gamma Multidimensional
Spectrometer for the Analysis of Low
Radioactivity Samples

N. A. Wogman

Many significant advances have been made in the development of low-level radionuclide analysis systems for non-destructive measurements of a complex spectrum of radioisotopes in an entire sample. Recently, a more selective system was required, and has been developed and applied for the analysis of radionuclides on the surfaces of environmental materials. The developed system utilizes two 30 cm diameter by 20 cm thick NaI(Tl) crystals with their associated 30 cm diameter by 10 cm thick NaI light pipes as the principal gamma-ray detection system. A 12.7 cm diameter by 1 cm thick P-10 gas-operated proportional beta detector with its 500 $\mu\text{g}/\text{cm}^2$ gold-plated Mylar window is placed between these two gamma-ray detectors, and the specimen to be analyzed is placed on the beta detector. This entire beta-gamma-gamma spectrometer system is shielded

from natural and cosmic radiation by a 4-inch thick lead cave. The two NaI(Tl) crystals record only those gamma-ray events occurring in coincidence with a beta particle. Gamma rays in coincidence with each other are stored in the energy-energy plane of a 64 by 64, 4096 channel memory according to their photon energies, while single gamma-rays which interact in either crystal are stored on the axis of the array. Since beta particles have a definite range in solid materials, events detected in the beta detectors must arise from surface phenomena. For example, ^{203}Hg , a neutron activation product of mercury, has a predominant maximum beta emission energy of 0.214 MeV. Since the range of this beta particle in tissue-like materials is about 0.25 mm, ^{203}Hg is detected only from this very thin surface layer. The range of the beta particle defines the depth from which the radionuclide gamma-ray events are recorded by the coincidence operation. The system also prevents electron capture radionuclides from being analyzed since these radioisotopes do

not trigger the beta detector. This system is being actively utilized to study neutron-activated trace elements in the surfaces of environmental materials such as bone and extra-terrestrial materials such as returned lunar specimens. Only with this technique can the deposition pattern of toxic trace materials be determined on and within bone. The utilization of this instrument is being expanded to determine deposition patterns of radionuclides on selected surfaces of environmental materials to define the deposition pattern of a specific radionuclide or trace element relative to the whole specimen.

Trace Element Content of Aquatic
Organisms from the Columbia River

L. A. Rancitelli and
C. E. Cushing, Jr.*

The major, minor and trace element content of six species of fish from the Columbia River were determined by instrumental neutron activation analysis. This was an effort both to establish the present levels of toxic heavy metals in the environment and to predict from their concentrations and the specific activities normally

encountered, the significance of radiologically important elements such as zinc and iron to these organisms. Samples of the dorsal muscle, liver and kidney from chiselmouth, squawfish, fine and coarse scale suckers, carp and sturgeon were analyzed for the following elements: K, Na, Rb, Cs, Zn, Fe, Br, Se, Cr, As, Hg, Co, Ag, Sb, Sc and Au.

Many of the trends in the elemental concentrations of tissues from Pacific salmon are also noted in the Columbia River fish such as the levels of K, Cs, Fe, Zn, Rb, Br, Se, Co and Ag in the muscle tissue. Silver was found to reside primarily in the liver as was the case for marine organisms. However, several significant differences in the elemental content of Columbia River organisms from those in Pacific salmon are also evident. While the mercury content of the muscle tissue from the Columbia River fish is much higher than Pacific salmon only two squawfish of the 25 fish sampled exceeded 0.5 ppm on a fresh weight basis. High concentrations of the radiologically important elements Zn and Fe were observed in the kidney and liver of all organisms. The sturgeon liver contained the highest iron content (8600 ppm) observed.

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Elemental Content of Columbia River Fish
(ppm dry tissue except as noted)

SPECIES, TISSUE	K(%)	Na(%)	Zn	Fe	Rb	Br	Se	Cr	As	Hg	Co	Cs	Ag	Sb	Sc	Au
CHISELMOUTH																
DORSAL MUSCLE	1.84	0.085	17	27	22	4.8	1.2	0.31	0.7	0.19	0.004	0.075	0.002	<0.0005	0.00012	0.13
LIVER	0.85	0.17	61	140	14	10	5.2	0.24	1.5	0.52	0.064	0.017	0.52	0.056	0.0014	0.0018
KIDNEY	1.04	0.32	310	360	31	16	3.1	0.57	1.6	0.37	2.7	0.038	0.004	0.0004	0.0001	0.0069
SQUAW FISH																
DORSAL MUSCLE	2.02	0.064	30	10	9.8	3.5	1.2	0.15	<0.18	0.26	0.003	0.016	<0.001	0.0008	0.00002	<0.0007
FINE SCALE SUCKER																
DORSAL MUSCLE	1.66	0.071	39	21	18	5.4	0.56	0.17	0.9	0.1	0.011	0.075	0.002	0.0007	0.00004	0.0011
LIVER	0.61	0.084	190	280	4.5	7.3	2.5	1.3	<0.4	1.1	0.076	0.003	0.49	0.021	0.0032	0.0013
KIDNEY	1.46	0.38	130	720	20	39	2.9	0.31	1.0	1.8	0.57	0.027	0.043	0.003	0.0008	0.0052
COARSE SCALE SUCKER																
DORSAL MUSCLE	1.70	0.081	37	21	17	5.6	0.92	0.19	0.52	0.20	0.012	0.075	<0.001	0.0028	0.00004	0.0009
CARP																
DORSAL MUSCLE	1.88	0.077	20	44	9.7	3.4	1.8	0.62	<0.5	1.1	0.020	0.029	0.0013	0.0004	0.00008	<0.0005
LIVER	2.56	0.33	160	360	30	42	4.8	0.32	1.7	0.49	0.17	0.019	0.12	0.016	0.0034	<0.0015
KIDNEY	1.04	0.35	920	630	1.2	29	9.5	2.0	<1.5	6.5	0.77	0.015	0.026	0.018	0.0073	0.014
STURGEON																
DORSAL MUSCLE	1.89	0.30	16	82	10	15	2.1	2.5	<1	2.5	0.24	0.060	0.0019	0.008	0.0017	<0.0006
LIVER	1.31	0.51	110	8600	17	31	13	0.66	5.4	3.8	0.96	0.013	0.90	0.14	0.022	0.0018
KIDNEY	1.96	0.48	140	600	12	36	5.8	0.45	4.5	0.9	0.25	0.017	0.012	0.011	0.0016	<0.0002

Elemental Composition of California Gull Eggs

L. A. Rancitelli and W. L. Templeton*

While biological systems are subjected to the deleterious effects of toxic elements and radionuclides throughout their life cycle, their sensitivity to these pollutants may be higher at specific stages of their development. The effects of DDT on the development of birds, for example, suggests that a particularly critical point in the life cycle is the egg development. Since pesticides can produce significant changes it is not unreasonable to expect that heavy metals or radionuclides present during egg development might also present a hazard to the species. For these reasons, the elemental content of California Gull eggs collected from nests along the banks of the Columbia River was determined by instrumental neutron activation analysis.

The concentrations of 13 elements, Ca, Na, K, Fe, Zn, Se, Br, Hg, Ag, Cs, Co, Sb and Sc, were measured in the shell, yolk, and albumen of 12 eggs. Of those elements which are radiologically important in the Columbia River ecosystem, Fe and Zn resided primarily in the yolk, Cs in the albumen and Co in the shell and yolk of the eggs. Scandium and antimony were rather uniformly distributed in all phases of the eggs. From the standpoint of heavy metal pollution, the mercury was found primarily in the albumen. On a fresh weight basis,

the average mercury content of the albumen was 0.5 ppm while several individual eggs contained as much as 1 ppm mercury.

Average Elemental Content
of California Gull Eggs
(dry tissue basis)

Element	Yolk	Albumen	Shell
Ca (%)	ND*	ND*	35.1 ± 1.7
Na (%)	0.26 ± 0.15	0.84 ± 0.22	0.16 ± 0.04
K (%)	0.46 ± 0.24	0.84 ± 0.37	ND
Fe (ppm)	150 ± 20	30 ± 5	32 ± 5
Zn (ppm)	69 ± 20	1.0 ± 0.2	4.0 ± 2.0
Se (ppm)	2.5 ± 0.7	3.8 ± 1.4	0.32 ± 0.04
Br (ppm)	32 ± 13	65 ± 38	7.4 ± 6.1
Hg (ppm)	0.19 ± 0.09	2.2 ± 1.2	0.05 ± 0.02
Ag (ppb)	<1 - 2.8	<1 - 1.6	<1 - 6
Cs (ppb)	1.8 ± 2.0	4.6 ± 2.9	1.9 ± 1.4
Co (ppb)	11 ± 4	2.6 ± 2.4	11 ± 18
Sb (ppb)	1.3 ± 1.0	2.2 ± 2.4	5.4 ± 2.8
Sc (ppb)	0.9 ± 0.7	0.2 ± 0.3	4.1 ± 3.8
Dry Tissue/ Fresh Tissue	0.35	0.23	0.92

* ND - Not Detected

The Performance and Application of
an Anticoincidence Shielded Dual
Ge(Li) Gamma Ray Spectrometer for
Low-Level and Gamma-Gamma Coincidence
Counting

J. A. Cooper, R. W. Perkins,
J. R. Kosorok and C. E. Jenkins

A high-efficiency anticoincidence shielded Ge(Li) gamma-ray spectrometer incorporating 70 and 80 cm³ high-resolution Ge(Li) detectors has been evaluated and applied to the measurement of radionuclides in bio-

* Ecosystems Department

logical and environmental materials. As a low-level gamma-ray spectrometer it is capable of measuring as little as 0.1 dpm of ^{137}Cs and 2 dpm of ^{60}Co . A 2 dpm source of ^{137}Cs can be measured with 10% accuracy in 1000 minutes, while the addition of 2000 dpm of ^{60}Co increases the ^{137}Cs error to only 20%. The system allows the measurement of radionuclides in environmental specimens at quite diverse concentration levels. In Chum salmon liver the radionuclides measured were ^{137}Cs -5 dpm, $^{108\text{m}}\text{Ag}$ -10 dpm, and ^{54}Mn -15 dpm at an accuracy of 10% even in the presence of 1000 dpm of ^{40}K . The $^{108\text{m}}\text{Ag}$ measurement made with this system represents the first instrumental measurement of this radioisotope in the environs providing a new tracer technique for long-term monitoring of the environment. In addition, air, water, vegetation and biological specimens have been analyzed for their environmental radionuclides in a routine manner. The system provides a significant improvement in sensitivity over previous anticoincidence shielded Ge(Li) gamma-ray spectrometers and complements the large Na(Tl) multidimensional analyzers for over two-thirds of the radionuclides encountered in environmental samples.

It has been demonstrated that Compton interferences are reduced about 10,000-fold when this system is operated as a gamma-gamma coincidence spectrometer. However, efficiencies are also decreased, so the increase in sensitivity is considerably smaller. Although the improvement in sensitivity is strongly dependent on the initial peak-to-background

ratio, sensitivity improvements of 10-fold are possible for radionuclides such as ^{75}Se , which emits low-energy coincident gamma rays.

Compton interferences due to coincidence gamma rays emitted in the decay of ^{60}Co are reduced 20-fold more with the anticoincidence shielding than without it. The anticoincidence shield is not as effective when the Compton interference is due to non-coincident gamma rays. The application of this technique to radio-analytical problems is limited to high-level samples such as found in many neutron-activated and fission product specimens. About 15 of these radionuclides have a potential for sensitivity improvement with this technique with those radionuclides emitting low-energy coincidence gamma rays and with half-lives longer than a few hours showing the greatest potential for significant improvements.

Technology Development for Dual Atmospheric Tracer Studies on an Instrumented Meteorological Grid

J. D. Ludwick

Nuclear techniques are useful in studies of air pollution problems including the dispersion of material from a source and its subsequent distribution throughout the environment. A noble-gas-tracer grid system has been developed and used in the quantitative measurement on a real time basis of the transport and diffusion of ^{85}Kr released as a puff or continuous source out to distances of 800 meters.

This developed ^{85}Kr monitoring system has been modified to allow continuous recording of real time data differentiated into 1 second time intervals. Each downwind detector was modified to include a remotely operated cover plate which can also be used as an absorber or energy discriminator between the ^{85}Kr beta particles and other airborne radionuclides. Thus, it is adaptable to the measurement of reasonably high and low energy beta emitters in the presence of ^{85}Kr (0.7 MeV). Typically, a high energy beta could originate from a radioactive particulate in the micron size range where unwanted effects from self-absorption would be negligible. Two techniques which have been considered for the generation of a 0.1 to 1 micron aerosol are the vaporization of a low-boiling solid material and the ignition of solid propellant-type materials.

Several factors must be considered in dispersing tracer radionuclides such as ^{85}Kr to the environment. The downwind radiation hazard must be reduced to a negligible factor. This can be accomplished by using least possible quantities of a short-lived radioisotope. In addition, the dispersal study must be conducted at a controlled location insuring that no detectable quantities of radiation leave the test site. For dual tracer experiments the beta energy of the particulate must be at least 50% dif-

ferent from that of the 0.7 MeV ^{85}Kr beta particle (or of the 0.35 MeV ^{133}Xe). Several radioisotopes prepared by neutron activation meet this criterion; these include ^{56}Mn , ^{31}Si , ^{64}Cu and ^{24}Na . Another convenient method for the preparation of such radioisotope sources would be in the use of a long-lived parent, short-lived daughter combination where a daughter of moderate strength could be separated from its parent at a time convenient to the field experiment needs.

The field experiments would be initiated by vaporization or ignition of the particulate tracer at the same time as the krypton vial burst. As the plumes traveled downwind, each monitor station's cover plate would be synchronously opened and closed at 1 second intervals. The thickness of the cover plate in the area over the detector surface would be adjusted to allow passage of only the harder beta particle. Thus data accumulated while the cover was open would represent the total air concentration of both the gaseous and the particulate tracer. During the closed cycle, however, only information from the harder beta tracer would be collected. In this way, not only would the unique real time particulate air concentrations be obtained but also, by comparison with the simultaneous noble gas concentrations measured, a quantitative measure of the depleting character of the particulate would be available.

Utilization of ^{252}Cf Neutrons for
the Measurement of Specific Activi-
ties in Environmental Materials

W. A. Haller and N. A. Wogman

Since trace elements and radionuclides do not behave similarly throughout an ecosystem, a method has been required to measure the specific activities in many environmental specimens. To date, these requirements have been fulfilled by the analysis of 1000-gram bulk samples for their radionuclide constituents followed by the irradiation of milligram aliquots of this material in a high-flux reactor. A primary problem encountered in this technique is that of sample inhomogeneity. A milligram specimen which is taken for neutron irradiation often is not representative of the bulk trace element. Therefore, the manmade spontaneous fission element ^{252}Cf , a neutron emitter, has been used to irradiate entire 1000-gram samples, thus providing a technique for the measurement of the specific activities of trace elements in the entire bulk material. This technique has been utilized to measure specific activities such as ^{65}Zn in Columbia River organisms. A 2-milligram ^{252}Cf source which provides a workable thermal neutron flux of 10^8 neutrons is also being used to irradiate 1000-gram aliquots of biological material for week-long periods. In this manner tissues which often fractionate trace elements can be analyzed for their entire trace element content. Specimens such as liver, lung and kidney can be analyzed for elements

such as Zn, Co, Se, Hg, Cr, Cs, As, Sc, Rb, Cl, Mg, Cu, Na, K and Br. These neutron-irradiated kilogram specimens are analyzed with multi-dimensional gamma-ray spectrometers or selective anticoincidence shielded Ge(Li) spectrometer systems. Since week-long irradiations are not practical as a standard analytical technique, a method of neutron multiplication at a 10- to 50-fold level is being considered for application to biological specimens.

Columbia River Water Trace Element
Analysis

T. M. Tanner and L. A. Rancitelli

Trace elements were analyzed in samples of Columbia River water taken at ecologically significant times in 1968 and 1969. These samples were taken in conjunction with phytoplankton sampling in a continuing effort to characterize Columbia River water, aquatic organisms and sediments. These analyses permit the tracing of concentrations of many biologically significant trace elements through the food chain to man. The analysis method by which 19 elements are determined employs neutron activation, a rapid group chemical separation and gamma-ray analyses.

Examination of the data indicates a seasonal variation in trace element concentration for many elements with peak concentrations occurring in the spring for Co, Cr, Cs, Fe, Hg, K, Mn, Na, Sb, Sc, Zn and U. Silver shows a very large unexplained peak in June

and Rb fluctuates from 0.4 to 1.3 ppb from February to August. Concentrations of As, Au, Br, Cu and Se were relatively constant throughout the year. Some elements, Ag, Au, Cr, Cs, Cu, K, Na and Rb show relatively high concentrations in specific samples. However, it is not known if this represents a peak in concentration since these elements were not determined before November, 1968.

For many elements, notably Ag, Co, Cr, Cs, Fe and Hg which exhibit concentration peaks in water, a similar increase is noted in phytoplankton which may suggest an increased uptake of these elements as they become available to the plankton. This study is being continued with samples of the caddis fly larva, the next step in the Columbia River food chain.

Variations in Columbia River Water Trace Element
Concentrations as a Function of Season
(parts per billion)

Element	11/14/68	2/13/69	3/12/69	4/24/69	6/11/69	8/14/69	Estimated Exptl. Error, %
Ag	0.50	<0.01	<0.01	<0.01	3.03	<0.01	20
As	2.5	1.4	2.5	2.9	2.6	2.4	10
Au	0.030	<0.005	<0.005	<0.005	<0.005	<0.005	20
Br	5.6	5.4	5.1	5.4	4.9	4.1	5
Co	0.02	0.04	0.84	0.22	0.06	0.09	10-20
Cr	0.4	<0.1	2.1	0.1	0.2	<0.1	25
Cs	0.35	0.03	0.12	0.02	0.03	0.03	25
Cu	2.6	0.6	1.4	1.5	1.0	1.1	5-25
Fe	13	10	30	62	24	17	10-20
Hg	<0.1	<0.1	<0.1	0.2	<0.1	0.1	20
K	1190	560	1060	1060	920	990	5
Mn	2.8	5.4	15.5	43.8	9.4	11.0	5
Na	2200	1365	2290	2900	1880	2100	5
Rb	3.5	0.4	1.0	0.9	1.1	1.3	25
Sb	0.24	0.43	0.71	1.04	0.30	0.43	10-20
Sc	0.002	0.001	0.003	0.015	0.003	0.006	20
Se	0.2	0.2	<<0.1	0.1	0.2	0.1	10-20
U	0.7	0.5	0.8	1.0	1.0	0.7	10-20
Zn	8	44	190	6	8	6	10-20

The Fate of Some Trace Elements in
Water Processed by Water Treatment
Plants

T. M. Tanner and L. A. Rancitelli

An analytical technique originally developed for the trace element determination of natural waters utilizing neutron activation, group chemical separations and gamma-ray spectrometry has been applied to the analysis of waters subjected to water treatment processes. Two types of water have been analyzed: one treated for the purpose of cooling a Hanford reactor with emphasis on the reduction of radionuclides in the reactor effluent, and the second, to produce drinkable water from an advanced municipal-type raw sewage treatment system.

In the former process, the river water is treated with a flocculation-filtration process using bauxite or liquid alum. Samples of untreated water and waters treated with the two types of flocculators were analyzed to determine what changes occurred in the processes and to what extent impurities were removed. Of the 18 elements determined in each sample, there was essentially no concentration change from raw water to treated water for the elements Br, Co, Cs, Fe, K, Rb, Sb, Sc, Se and Zn. Both flocculation processes reduced the As by fourfold and U by twofold. Manganese showed a slight reduction

in the liquid alum process and a larger reduction by the bauxite process. Copper showed an almost sevenfold reduction by both processes from raw water. Silver was removed by the bauxite process; however, it appears that Ag may be a contaminant in the liquid alum. Sodium and chromium show increases though not in the same proportions in both processes which may be due to the addition of sodium dichromate as a corrosion inhibitor. Mercury is seen to increase by greater than threefold in both processes and is probably due to contaminants in the liquid alum and bauxite.

The second type of waters examined was from an advanced treatment system in South Lake Tahoe, California, which processes raw sewage into drinkable water. Correlation of the water treatment process and the data indicate that the samples may be divided into two similar fractions: raw sewage and secondary effluent, and filter-final effluent. There are five elements which were significantly reduced in concentration: Co by threefold, Cu about 30 percent, Mn by thirtyfold, U by fourfold and Zn by fourfold. Arsenic, Cs, K, Na and Sc showed essentially no change and the remaining elemental concentrations showed net increases through the process which were due to chemical additions.

The sensitive multielement capability demonstrated in the analysis of these two different types of processed waters may become increasingly important as our knowledge of the biologically and ecologically significant levels of trace elements increases. In this respect two uses of

this method become immediately obvious: the monitoring of processed water with respect to trace elements at very low levels; and, after trace element characterization of a process plant's effluent, tracing the effluent plume as it re-enters the water supply.

Elemental Concentrations in Columbia River Water Treated
by Liquid Alum and Bauxite Processes
for Use as Reactor Coolant Water
(parts per billion)

Element	Raw Water	Liquid Alum Treated	Bauxite Treated	Error, %
A	0.13	0.94	<0.01	20
As	1.4	0.3	0.3	10
Br	4.9	3.8	3.6	5
Co	0.02	0.03	0.01	10-20
Cr	<0.1	0.2	0.5	25
Cs	0.01	0.02	0.02	25
Cu	14.3	2.6	2.3	10-20
Fe	<15	<15	<15	20
Hg	<0.1	0.3	0.3	20
K	970	920	880	5
Mn	2.2	1.9	1.3	5
Na	2150	2920	2540	5
Rb	1.0	1.3	0.9	25
Sb	0.24	0.24	0.17	10-20
Sc	0.001	0.001	0.001	20
Se	0.1	<0.1	0.1	10-20
U	0.8	0.3	0.4	10-20
Zn	28	27	23	10-20

Neg 706013-1

A Beta-Gamma-Gamma Multidimensional Spectrometer System Provides A Totally Nondestructive Means to Determine the Surface Concentrations of Beta Emitting Radionuclides or of the Coincident Gamma Rays from Neutron Activation Products.

Neg 706013-2

Neg 704620-3

Anticoincidence-Shielded Dual Ge(Li) Gamma-Ray Spectrometers
Are Providing the First Direct Measurement Technique
to Determine the Radionuclide ^{108m}Ag Which May be Utilized
to Trace Elemental Silver in the Environment and Relate
Possible Ecological Concentration Factors
to Environmental Conditions.

GAMMA-RAY SPECTRA OF ^{54}Mn AND ^{75}Se

Neg 705062-1

Compton Interferences Are Reduced Four Orders of Magnitude
Using Gamma-Gamma Coincidence Ge(Li) Spectrometry
on a Mixture of ^{54}Mn and ^{75}Se Produced by Fast-Neutron
Activation Providing an Increased Sensitivity
in the Measurement of ^{75}Se .

TRACER STUDIES IN THE NORTHEAST COLORADO
HAIL EXPERIMENT PROGRAM

Studies of the cloud dynamics and microphysics of severe convective storms are being made through measurements of cosmic-ray-produced radionuclides, trace elements, and added inert or radioactive tracer elements to identify effective means of modifying those storms that produce damaging hail fall and to determine whether the chosen modification methods are technically and economically feasible.

Atmospheric Tracer Techniques for the
Northeast Colorado Hail Experiment

N. A. Wogman and J. A. Young

Unique atmospheric tracer techniques utilizing cosmogenic and specific tracer elements have been developed which are providing methods to characterize specific atmospheric processes. Preliminary background information has been obtained for the four primary experiments which will be conducted during the Northeast Colorado Hail Experiment (NECHE). These experiments are (1) the measurement of the short-lived cosmogenic radionuclides ^7Be , ^{24}Na , $^{34\text{m}}\text{Cl}$, ^{38}Cl , and ^{39}Cl in hail and associated rain during consecutive sampling periods of Northeast Colorado convective hail storms, (2) the characterization of the trace element composition of the precipitation, (3) the evaluation of the release, collection, and analytical procedures for the air mass tracers Ir, Ru, Au, Te, Se, In, and Ag, and (4) the determination of the effectiveness of seeding with Ag^{129}I to produce ice nuclei.

Air filter samples were collected for trace element analysis in the NECHE area utilizing an RB-57F aircraft as a collection platform. The atmospheric concentrations of the above tracers and also Fe, Zn, Cr, Co, Sb, and Sc were determined. These measurements indicate that it will be possible to increase the atmospheric concentrations of Ir, Ru, Au, Te, Se, In, and Ag by a few orders of magnitude through their release in pyrotechnique flares.

The trace element composition of hail or rain should bear a definite relationship to the initial nucleation and subsequent in-cloud processes to which the particle was subjected. The measurement of the variations of the background trace element and the added tracer compositions of hail and rain with hailstone size will establish any relationship existing between the ice nuclei and total aerosol concentrations in the air and the hailstone size. These background measurements were initiated by analyzing hailstones obtained during 1970 by a tech-

nique of neutron activation analysis. Trace element concentrations of elements such as In and Au range from 1 to $<5 \times 10^{-11}$ g/ml of hailstone and are of the same magnitude as earlier precipitation concentrations measured in rain from Quillayute, Washington. The insertion of the nonnucleating tracers Ir, Ru, Au, Te, Se, and In at discrete positions around the convective cloud followed by their subsequent measurement in precipitation will provide information on the movements of air, rain, and hail within the cloud. The seeding of the cloud with Ag^{129}I at one location and with ordinary AgI at another will provide information on the relative efficiencies of seeding at the two sites.

The hail formation process can also be studied using cosmogenic radionuclides. These radionuclides are produced at a constant rate in

the atmosphere and quickly become attached to the atmospheric aerosol, so they can serve as tracers of the aerosol particles. After the aerosol particle becomes part of a droplet or ice particle, the cosmogenic radionuclides decay at rates which differ because of their different half-lives. The ratio of two cosmogenic radionuclides in precipitation can therefore be used to calculate the time since the aerosol was incorporated in the precipitation particle. The $^{39}\text{Cl}:^{38}\text{Cl}$ ratio will be determined as a function of the depth within the hailstone to determine the time between the condensation of the water or ice and the arrival of hailstones at the ground. The concentrations of the cosmogenic radionuclides will also be used to calculate scavenging efficiencies of the precipitation.

Neg 702865-2

Cosmogenic Radionuclide-Tagged Aerosols Provide Tracers
of Atmospheric Precipitation Processes

RADIOLOGICAL PHYSICSINTERNAL DEPOSITIONS OF RADIONUCLIDES IN MAN

These studies are concerned with the continued development and improvement of diagnostic techniques for the assessment of internal depositions of radiotoxic materials in man and the development and improvement in dosimetry associated with such depositions.

Promethium

D. McConnon

Efforts continue toward development of a diagnostic method for inhaled $^{147}\text{Pm}_2\text{O}_3$. An experiment was performed (by J. E. Ballou, Inhalation Toxicology Section, Biology Department) with rats to determine whether there are any gross differences in the metabolism of samarium and promethium. Promethium-147 and ^{145}Sm chlorides in a citrate buffer solution at pH 4.5 were injected intravenously into two groups of nine rats each. One group was injected with 4 μCi of ^{147}Pm per animal while the other group received 3 μCi of ^{145}Sm per animal. These animals were sacrificed 14 days after injection and samples of bone, liver, kidney, muscles, etc., were analyzed for the appropriate radionuclide. Translocation and excretion patterns were followed with samples of blood, urine and feces. From the results of this experiment, it is concluded that no

gross differences occur in the metabolism of samarium and promethium in the rat. The favorable results of this experiment allow continuation of the program into the next phase involving inhalation by dogs of an aerosol similar to that planned for the human experiments.

Efforts continued on producing an aerosol from a samarium-promethium oxide suspension that would be suitable for use in human testing. A water suspension of Sm_2O_3 calcined at 1110°C was prepared for particle size studies. The aerosol generated from this suspension had a mass median aerodynamic diameter (MMAD) of $0.85\ \mu\text{m}$ with a geometric standard deviation of 4.2. The observed MMAD is in a range suitable for the human studies; however, the large standard deviation indicates that the aerosol is extremely heterodisperse thus necessitating treatment of the oxide suspension prior to aerosol generation to remove the larger particles. It was found that a simple settling

procedure in which the sample is allowed to remain undisturbed for a time eliminated most of the larger particles. The mass of material that is lost depends upon the calcining temperature. Data on the effect of calcining temperature on particle size distribution are not yet complete, but preliminary indications are that the particle size distribution increases with higher calcining temperatures.

The ^{143}Pm used for the human injection experiments was produced by a (p,2n) reaction on highly enriched ^{144}Nd . Because the final product contained approximately 10% ^{144}Pm by activity due to the competing (p,n) reaction, it was decided to attempt a production run by another means to reduce the amount of ^{144}Pm present. Promethium-143 made according to the following production path

was ordered from the Oak Ridge National Laboratory. Separation of the product from the samarium target was done at ORNL and a 10 ml sample containing approximately 210 μCi of ^{143}Pm was received. Measurements with a high resolution Ge(Li) detector indicated very pure ^{143}Pm with no evidence of a gamma-emitting contaminant. The samarium content of the sample

was less than 50 ppb as measured with a spark source mass spectrograph indicating a very clean separation of the product from the target material. Alpha analysis of the sample by 2π alpha proportional counting revealed no alpha-emitting contaminants. The importance of production of ^{143}Pm without ^{144}Pm is illustrated by the fact that the calculated doses to selected critical organs will be increased by an average of about 44% with ^{143}Pm that contains 10% ^{144}Pm by activity.

To assess the feasibility of establishing the amount of ^{147}Pm in samples through measurement of the 121.2 keV gamma ray emitted from an excited level of the daughter ^{147}Sm , a measurement of the disintegration yield or emission probability was made. Two such measurements using two different reference sources yielded values of $2.90 \pm 0.23 \times 10^{-5}$ and $2.95 \pm 0.17 \times 10^{-5}$ for a weighted average of $2.93 \pm 0.14 \times 10^{-5}$ gamma rays per disintegration.

A study of the feasibility of estimating depositions of aged ^{147}Pm in humans by whole body counting techniques disclosed that a minimum detectable amount between 0.5 and 1.0 μCi , depending upon the chest wall thickness, could be attained using the lower energy bremsstrahlung peak. These results indicate that with specialized whole body counting equipment, the minimum detectable amount of ^{147}Pm measurable in the lungs is on the order of 10 to 25% of a maximum permissible lung burden (12 μCi).

Plutonium

I. C. Nelson and D. McConnon

Interest in development of a diagnostic method to be used in the presence of chelating agents used for therapeutic removal of plutonium continued; however, the processes utilized in preparation of test quantities of ^{237}Pu have not yielded products of sufficient purity for use in human volunteer test subjects. Our experience suggests that the next attempt at production of pure ^{237}Pu may require use of isotope separation on alpha-irradiated ^{235}U or a fresh start on cyclotron irradiation of neptunium. Either of these approaches would be expensive in terms of present funding and further efforts on production of ^{237}Pu were postponed.

The autopsy program was continued. Laboratory facilities were readied and processing of samples obtained during 1968 to present initiated. Completion of laboratory and dosimetric analysis and publication of a 5-year updating report is planned for 1971.

Radon Daughter Deposition in the Respiratory Tract

I. C. Nelson and B. O. Stuart*

Lung cancers in uranium miners are thought to originate predominantly in

the hilar region of the lungs. The role of alpha-emitting radon daughters prevalent in the mines in contributing to the increased incidence of these cancers has not been well-defined. One major uncertainty in estimating the dose to the target tissue from these alpha particles is the deposition of the radon daughters on the tracheobronchial mucosa. The radioactive material is apparently carried to the respiratory tract as unattached ions, on condensation nuclei and on larger dust particles. In this study, uranium mine aerosols are passed through a hollow RTV rubber cast of a teaching model of the human tracheobronchial tree. The tree is then cut up into identifiable units and the deposited activity measured. The apparatus and technique are presently being perfected; however, preliminary results indicate that the areal deposition of activity in the bronchi relative to that in the trachea increases by a factor of about 2 in the lobar bronchi and then diminishes in the segmental bronchi. Plans include treatment of the hollow cast with mucus-like substances to improve similarity to the human airways, use of a variety of airflow patterns and pollutant mixtures, preparation of casts from cadavers to assess importance of biological variability in airway structure and similar experiments on models of the human nasopharynx and larynx.

* *Inhalation Toxicology Section,
Biology Department.*

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Hollow RTV Cast (Right) of Teaching Model of Tracheobronchial
Tree (Left) Used in Uranium Mine Aerosol Deposition Studies

Lung Dosimetry of Radon Daughters

I. C. Nelson

Theoretical development of the dose to the tracheobronchial epithelial basal cells has included estimates based on diffusion theory and the Weibel regular dichotomous lung model. The conductive airways in the real lung can be better described by irregular dichotomy which departs from the regular model principally in lengths of bronchi and bronchioles. The actual lengths and diameters found by Weibel in the preparation of his model were obtained from him for branches through the seventh generation. Airway surface areas, cross sections and volumes were calculated preparatory to calculation of areal deposition of stylized aerosols containing radon daughter activity using diffusion theory. Air flow rates and tube dimensions realized in life are such that present mathematical developments of diffusing particles are not considered strictly applicable; however, these calculations will establish the relative usefulness of the simplified, regular dichotomous model over the more complex irregular dichotomous model.

The relative importance of flow patterns in estimating deposition by diffusion was examined assuming that inspiration and expiration followed a sinusoidal rather than a simplified square wave airflow pattern. For a given respiratory volume and time, calculations employing diffusion theory suggest an increase in deposi-

tion by a factor of about 3 for sinusoidal flow indicating a need for further consideration of this parameter in detailed calculations.

Estimates of the cancer-related dose also include estimates of the alpha particle energy deposited in the nucleus of the epithelial basal cells. The development of the mathematical geometry, LET and energy deposition after the method of Haque is being examined in detail to establish the importance of simplifications made in these parameters in establishing the relevant dose.

PUBLICATIONS

D. McConnon. "The Use of Solid State Detectors for Organ Counting," BNWL-1178. Battelle-Northwest, Richland, Washington, May 1970.

D. McConnon. "The Use of Neptunium-235 for the Calibration of Plutonium Lung Counters," BNWL-SA-3191. Battelle-Northwest, Richland, Washington, 1970. To be published in International Journal of Applied Radiation and Isotopes.

D. McConnon. "Measurement of the γ Disintegration Yield in the Decay of ^{147}Pm ," BNWL-SA-3560. Battelle-Northwest, Richland, Washington, 1970. To be published in International Journal of Applied Radiation and Isotopes.

D. McConnon. "Radiation Types and Intensities in the Decay of ^{235}Np ," BNWL-SA-3453, Battelle-Northwest, Richland, Washington, 1970. To be published in Health Physics.

I. C. Nelson, "Build-Up of Americium-241 in Lung Containing Plutonium-241," BNWL-B-12. Battelle-Northwest, Richland, Washington, September 1970.

CHARACTERIZATION OF RADIATION-INDUCED FREE
RADICAL REACTIONS IN AQUEOUS SYSTEMS

R. N. Diebel, W. D. Felix and D. R. Kalkwarf

This project is concerned with determining the chemical structures and reactivities of free radicals formed in irradiated model systems in order to understand and alleviate the development of radiobiological damage. Electron spin resonance spectroscopy and optical absorption spectrophotometry have been used to identify the radicals and determine the kinetics of their reactions with other biochemical substrates in aqueous solutions. The damage-propagating abilities of the radicals have been assessed by studying their reactivities toward diagnostic reagents, while methods for chemical protection from free-radical damage have been tested in various biochemical systems.

Several preparative methods were utilized to determine the structures and reactivities of radiation-induced biochemical radicals. Formation of radicals in gamma-irradiated crystals continued to be a convenient method for stabilizing them for structural and quantitative analysis prior to their dissolution in aqueous solutions of diagnostic reagents. Bleaching of eriochlorine at pH 6 and of p-nitrosodimethylaniline at pH 9 were used to detect the presence of oxidizing radicals, while one-electron reduction of flavin mononucleotide in 2N sulfuric acid to its stable-radical form was used to detect reducing species. Instrumentally broadened ESR spectra of the irradiated solids were measured to accurately determine the concentration of radicals by the first-moment method, while high-resolution spectra were also recorded to help discern their chemical structures.

Aliphatic amino acids, with the exception of cysteine and glutamic acid, were found to form oxidizing radicals; and, generally, the most potent oxidants were anionic or zwitterionic radicals in which the positively and negatively charged groups were not widely separated. Thus, CH_2COO^- formed in the γ -crystalline form of glycine and $\text{CH}_3\dot{\text{C}}(\text{NH}_3^+)\text{COO}^-$ formed in α -alanine were found to be potent oxidants, while $(\text{CH}_3)_3\dot{\text{N}}^+$ formed in betaine and $\text{H}_3\text{N}^+\text{CH}_2\dot{\text{C}}\text{HCOO}^-$ formed in β -alanine were unreactive in all tests. While this variety of behavior may reflect real differences in oxidation potential of the radicals, lack of attack on the reagents may also be due to rapid radical recombination or disproportionation reactions. The simple sugars, arabinose and galactose, as well as polysaccharides derived from them were also found to yield extremely potent oxidants; however, no structural

reason is yet apparent. Many sugars and molecules containing hydroxyl groups (e.g., mannitol and serine) formed radicals which reduced flavin mononucleotide; however, notable exceptions were found (e.g., sucrose, fructose and the natural nucleosides). Again, structural reasons for this variation in reactivity must await future experiments.

Biochemical radicals were also produced by the attack of photochemically generated hydroxyl radicals on various substrates. Neutral solutions of amino acids containing 0.1M hydrogen peroxide were illuminated with a 150-watt sealed beam xenon arc as they slowly passed through the sample cell of the ESR spectrometer. The ESR spectra indicated that hydrogen-atom abstraction from a methyl or methylene group was the predominant process with $\dot{\text{C}}\text{H}_2\text{CH}(\text{NH}_3^+)\text{COO}^-$, $\text{OOC}\dot{\text{C}}\text{HCH}_2(\text{NH}_3^+)\text{COO}^-$, and $\text{HO}\dot{\text{C}}\text{H}(\text{NH}_3^+)\text{COO}^-$ being obtained from α -alanine, glutamic acid, and serine, respectively. Solutions of phenylalanine and tryptophan in hydrogen peroxide also showed ESR spectra during photolysis, but there was insufficient resolution for a structural determination. In all cases, the radical signals disappeared "immediately" when illumination ceased.

Flash photolysis techniques were further developed in order to generate extremely transient radicals in sufficient concentrations for structural

and kinetic investigations. Using a high intensity flash lamp developed in this laboratory, decay kinetics of hydroxyl radicals in pure aerated water are being studied by ultraviolet spectrophotometry. In conjunction with these and other free-radical reactions, a quantum-mechanical computation technique is being developed to predict the rate constants of free radical reactions from their structural and energetic parameters. The technique is based on the CNDO molecular orbital method, suitably modified for molecules with unpaired spin. Correlation between computed and experimentally determined rate constants for these reactions should provide a more generalized basis for assessing free-radical activities as functions of chemical structure.

PRESENTATIONS

D. R. Kalkwarf. "Propagation of Radiation Damage with Biochemical Radicals," Fourth International Congress of Radiation Research, Evian-les-Bains, France, June 1970.

W. D. Felix. "High Intensity Z-Pinch Plasma Lamp for Flash Photolysis," Northwest Regional Meeting of the American Chemical Society, Seattle, Washington, June 1970.

R. N. Diebel. "Amino Acid Radicals Produced by Ultra Violet Photolysis of Solutions Containing Hydrogen Peroxide," Northwest Regional Meeting of the American Chemical Society, Seattle, Washington, June 1970.

NUCLEAR TECHNIQUES IN MEDICAL SCIENCE

The purpose of this study is to perform research in the application of nuclear technology and radioisotopes to the fields of medicine and biology and to combine the advanced knowledge, techniques, and instrumentation of the nuclear field with the talent and knowledge of the medical profession to improve and increase the direct medical benefits to mankind.

In-Vivo Activation Analysis

H. E. Palmer and K. G. Pailthorp

with

W. B. Nelp, J. D. Denney, R. Murano,
T. G. Rudd, J. L. Williams,
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Total Body Calcium Measurements

Using the in vivo activation analysis method for total body calcium which has been developed during the last few years,⁽¹⁾ over 100 measurements have been made on patients suffering from metabolic bone disease. Many of these patients have had repeat measurements to study serial changes in bone mass caused by a variety of diseases. The largest group of patients is under study for renal osteodystrophy and are maintained periodically on artificial kidney hemodialysis. Repeat measurements at approximately 6 and 12 month intervals after the first measurement were made on some of these patients. In two patients the measurements are essentially the same, but in the others there was a consistent trend of small calcium loss. One patient with

renal failure on hemodialysis showed a calcium loss of 14% (167 grams) during a period of 12 months. Lower levels of total body calcium were observed in patients of various disease groups which included osteoporosis, multiple myeloma, and renal transplant when compared to a group of normal subjects. The lowest calcium content was measured in those having osteoporosis.

To more accurately determine absolute amounts of total body calcium, three additional cadavers were accurately irradiated and counted several times and then carefully ashed by slow heating in a large muffle furnace this year. Chemical analysis of the ash is presently being done.

Total Body Sodium Measurements

Total body, exchangeable, and non-exchangeable sodium were determined on five normal adults using a combination of in vivo activation analysis and isotopic dilution. The average total body sodium was found to be 49 meq/kg and the nonexchangeable fraction ranged from 22 to 33% with an average of 27%.

Serial Loss of Body Calcium for Renal Osteodystrophy
Hemodialysis Patients as Determined
by In Vivo Activation Analysis

Total Body Calcium Measurements for Patients
in Various Disease Groups

Total Body Phosphorus in Mice

It appears feasible that total body phosphorus in humans can be measured more accurately with a much lower radiation dose than that required for total body calcium. To gain familiarity and confidence with the 14 MeV $^{31}\text{P}(n,\alpha)^{28}\text{Al}$ reaction and to determine if extra-skeletal phosphorus has a constant relationship with total body potassium under conditions of potassium or phosphorus deficiency, 21 mice were analyzed for phosphorus by in vivo activation analysis and subsequently by ashing and chemical analysis. The results of both methods of analysis show good agreement with a correlation coefficient of 0.94. The precision of the neutron activation method was $\pm 1.0\%$ and the accuracy was 3%.

Comparison of Chemical Versus
Activation Analysis
Determination of Phosphorus
in Mice

A Counter for the Direct Measurement
of ^{55}Fe in Blood and Other Liquid
Samples

H. E. Palmer

A counter was developed to count ^{55}Fe directly and rapidly in blood or liquid samples. This development reduces the required analysis time from days to minutes with only a small reduction in sensitivity when compared to other methods.

The unique feature of this counter is the sample holder. One hundred fifty feet of polyethylene tubing are coiled onto a Lucite frame. The tubing has an inside diameter of 0.023 in. and a wall thickness of 0.0075 in. The tubing is divided into lengths each 50 ft long and having a volume of about 4 ml. The three sections of tubing can be filled simultaneously or separately to give a choice of sample size 4, 8, or 12 ml. The tubing is charged with the liquid sample under a pressure of 20 lb/in.². The tubing easily withstands this pressure, and charging times are 1 minute for a dilute water solution and about 3 minutes for a more viscous blood sample.

For counting, the sample holder is placed between two large-area, thin window proportional counters. The upper and lower sections of the proportional counter each consists of a main counting chamber made up of four $2 \times 2 \times 16 \frac{1}{2}$ in. counters each separated by a grid of ground wires. A ring of $1 \times 1 \times 16 \frac{1}{2}$ in. anticoincidence guard counters surrounds the main chambers except for

the entrance side which faces the opposite counter. The anticoincidence guard chambers are separated from the main chambers by a grid of 0.001 in. wire spaced 1/8 in. apart. The thin entrance windows are made from 0.00015 in. thick aluminized Mylar. The counters are constructed from 1/4 in. thick Lucite, painted on the inside with colloidal graphite to provide a conducting surface. The ends of the counter are not painted and are nonconducting. A mixture of 90% argon, 10% methane gas flows through the counter at atmospheric pressure during operation. The counter has excellent resolution and a low background when operated inside a 2 in. thick lead shield and in anticoincidence with the guard chambers. The background is three counts per minute in the energy region of 4.6 to 7.2 keV. Twenty-five percent of the X-rays emitted in an ^{55}Fe solution contained in the tubing interact with the counting gas. The 2 in. thick main chambers absorb about 93% of the X-rays which come through the entrance windows.

The determination of ^{55}Fe in non-liquid biological samples can be accomplished by ashing the sample, dissolving the ash in dilute nitric acid and counting this solution in the counter. Acids containing heavier elements such as chlorine, sulfur, phosphorus, etc., must not be used since they strongly absorb the 5.9 keV X-rays.

This counter has been in use for several types of blood and liquid samples for about 1 year. The background and sensitivity have remained constant. In addition to the time

saving and simplicity feature of this method, it should be inherently more accurate since the measurement does not depend on a constant chemical yield or recovery as do the methods involving chemical separation and electrodeposition. In addition to the extensive medical uses of this method, the counter is sensitive enough to measure ^{55}Fe from nuclear fallout in the blood of Alaskan caribou, Eskimos, and other population groups.

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Sample Holder for Counting ^{55}Fe
in Blood and Liquid Samples

Radiation Instrumentation

K. L. Swinth

This task focuses on the development of advanced instrumentation concepts, detectors, and circuitry, and on the determination of their feasibility for use in the detection and measurement of ionizing radiation with emphasis on the measurement of in vivo depositions of alpha emitters such as plutonium.

Detection of plutonium translocated in the bronchial lymph nodes progressed through better definition of counting parameters and with animal and phantom evaluation of developed probes. Plutonium translocated to the bronchial lymph nodes can be detected by X-ray counting probes placed nearby in the esophagus. Data collected from the literature and from observation of cadavers improved the definition of the counting geometry involved in the detection of plutonium in bronchial lymph nodes. This indicated that the expected sensitivity would be better than anticipated from our original assumptions concerning the geometry; however, it also indicated that large errors would be inherent in the measurements due to variability of geometry. Quantitative measurements are not available to assess the magnitude of these errors.

Experiments were performed by placing the probe in both the esophagus and in the trachea of a number of dogs. (An X-ray photograph shows the position of the probe in a dog esophagus.) Postmortem experiments were also performed. Data from the dogs indicated a sensitivity of

76 counts/min/ μ Ci for a 1 minute count. Improvements in probes should lower this MDA below 13 nCi. By movement of the probes in the esophagus, a definite peak in the activity was shown to exist in the region of the lymph nodes.

Fabrication of a human chest phantom allowed evaluation of the probes in a realistic laboratory geometry. The chest phantom is temperature controlled and has a geometrical relation of esophagus, lymph nodes, and trachea similar to those in an average man. With this phantom, MDAs of 7 nCi to 22 nCi were obtained with a 20-minute counting time. The MDAs vary with assumptions concerning the exact geometry.

A number of experiments were performed to delineate the physical operating parameters of the probes. Temperature sensitivity, angular sensitivity, stability and background rates were measured. Improvements in detectors suggested by the experiments performed will improve the detector sensitivity and lessen geometrical variability.

A laboratory fabricated NaI(Tl) scintillation system with a fiber optics light guide was tested and found useful for detection of plutonium in the bronchial lymph nodes. This prototype had an MDA of about 50 nCi, comparable to a value of 25 nCi for the avalanche detector. The scintillation assembly depends primarily on detection of the ^{241}Am gamma rays (60 keV) and exhibits an efficiency of 1% for 17 keV X-rays. Although the gated background is over two orders of magnitude higher than for the avalanche

probes, due to tube noise and Cerenkov radiation, an order of magnitude reduction is obtained by pulse shape gating. It is expected that optimization of system parameters could substantially reduce the background and increase the sensitivity.

X-ray-alpha measurements were completed for a number of transuranic isotopes. For ^{242}Cm , ^{244}Cm , and ^{237}Np respective values of 0.117 ± 0.003 , 0.0918 ± 0.0013 , and 0.597 ± 0.006 were obtained using a solid-state coincidence system. These values along with results for plu-

tonium isotopes will prove helpful in the health physics interpretation of L X-ray counting results. Plutonium photon intensity data are being accumulated for the plutonium intercalibration studies following a meeting of those interested.

Wound visualization studies progressed with construction of an optics system for the image intensifier system and with the design of a position-sensitive proportional counter. Final testing of the systems awaits arrival of necessary accessory equipment.

X-ray Showing Avalanche Detector Positioned in Esophagus to Count the Uranium L X-rays from Plutonium Located in the Bronchial Lymph Nodes. (The bifurcation of the trachea is just below the detector.)

Variation in Count Rate with Position as an Avalanche Detector
Is Moved down the Esophagus Past the Lymph Nodes

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RADIATION DOSIMETRY

These studies are primarily concerned with the stochastic nature of absorbed dose. The objective is to determine those features of energy deposition patterns that are relevant to the understanding of observed biological effects. Theoretical effort has been in development of mathematical models and computational techniques to couple the spatial distributions of energy deposition (microdosimetry) with temporal distributions (hit theory). Experimental work has produced distributions of ionization in tissue-like sites for monoenergetic protons; emphasis has been on energy transport by secondary electrons.

Ionization Distributions

W. A. Glass and W. C. Roesch

Measurement was completed of the ionization distributions produced by fine beams of protons in wall-less proportional counters which simulate small biological sites. Our previous work had shown the necessity of using wall-less counters to simulate correctly the contributions due to secondary radiations. After developing methods of centering the counter in the beam and calibrating the system, a definitive set of data was obtained for 1.7, 3.0 and 4.0 MeV protons.* Two site sizes were simulated, a cylinder $25 \mu\text{g}/\text{cm}^2$ high and $25 \mu\text{g}/\text{cm}^2$ in diameter and another 100 by 100 $\mu\text{g}/\text{cm}^2$.

* One of us (WAG) is grateful to the Radiological Research Facility of Columbia University and the Brookhaven National Laboratory, and especially to H. H. Rossi, for their hospitality and for the use of their accelerator to obtain the data.

One of the reasons for the study of these ionization distributions was to understand better the role of secondary radiations on biological sites. Effects of secondaries were apparent by the presence of distributions for protons that passed by outside the site ($r \geq 12.5 \mu\text{g}/\text{cm}^2$); in such cases, all the ionization produced in the site is due to secondaries. In practical irradiations, the site is uniformly exposed to a broad beam rather than to the narrow beam used for the individual distributions. Distributions for a uniform beam were constructed theoretically from the data. In addition, it was possible to calculate the contributions to the total by protons that pass outside and by those that pass through the site. On the basis of our previous work, it is clear that experiments with solid-wall counters could not show the contribution due to protons passing outside the site. Experiments with wall-less counters have

shown the total curve but not the resolution into contributions from inside and outside events. For smaller sites, the relative contribution of the secondaries will be larger and the effects of secondaries increase with proton energy. These results thus give insight into the role of the secondary radiations.

Our theoretical work has shown the relationships between the different distributions. They can be understood as examples of a compound-Poisson process. The differences in shapes of the distributions are due to differences in the number of atomic collisions of the proton that can contribute to the ionization and to differences in transport of energy by secondary radiations. Study of statistical parameters such as the means and variances of the distributions indicated that most of the distributions due to protons that passed through the site, but at different distances from the center, were only

slightly affected by changes in the secondary energy-transport and that, therefore, they should be related through the mathematical process known as folding (or convolution). The relation could not be demonstrated by the use of the folding operation itself, because a non-integral number of steps would be involved; however, it was possible to use Fourier transforms for the purpose and to show how to pass from one distribution to another.

To provide a data bank for use in analysis of radiobiological phenomena, the large amount of data obtained in this work was normalized, averaged, gain-adjusted, smoothed, and interpolated and extrapolated to provide families of curves giving the ionization distributions for regularly spaced intervals in the ionization and in the distance from the center of the site for all ionizations and distances likely to be of any significance.

Ionization Distributions for 3 MeV Protons Passing the $25 \mu\text{g}/\text{cm}^2$ Absorbing Site at Various Distances from the Center

Ionization Distribution for Broad Beam Irradiation of a $25 \mu\text{g}/\text{cm}^2$ Absorbing Site as Composed from Individual Distributions. [Curve (a) is the total energy deposition spectrum, curve (b) is the contribution from protons passing outside the site, curve (c) is due to inside events.]

Hit Theory

W. C. Roesch

An extension of hit theory which was helpful in explaining radiobiological effects due to repair or recovery⁽¹⁾ made use of some approximations of limited accuracy in the calculations that were reported. These calculations have been improved. The calculations were of the probability, P_n , of surviving n hits. For the numerical work, it was assumed that the probability of the consequences of two given hits producing lethality decreased exponentially with the time between the hits.

An exact mathematical expression for P_n can be given; it is a multiple integral of order n of a large number of factors representing the interaction probabilities between hits. It proved to be very time consuming to evaluate. Using an n -dimensional Simpson's Rule, integration took prohibitive times for n greater than 3. The multiple integral is amenable to the Laplace transform technique, but the number

of terms involved leads to frequent errors. Therefore, a computer program was developed that does both the algebra of finding the Laplace transform and the numerical work of calculating the result from it. It was faster than the Simpson integration, but it too became prohibitive for n greater than 5. Although limited in the range of n for which they were applicable, these exact methods have been very useful in providing comparisons for faster, approximate calculations.

A Monte Carlo method of evaluating P_n was prepared which agreed within 1% with the exact methods. A number of approximate methods were investigated. The most accurate was one in which the n -dimensional integral of factors was replaced by those factors evaluated at their individual means. Survival curves computed from this approximation for several values of the parameters entering the calculation compare satisfactorily with values obtained by the Monte Carlo method. Use of the approximation should permit more extensive use of the theory.

Comparison of Survival Curves Calculated by Approximate
Evaluation of an n-Dimensional Integral with Values
Calculated by Monte Carlo Method

Heat Defect Calorimetry

D. M. Fleming

The heat defect calorimeter was re-designed resulting in improved accuracy and, more importantly, the time of exposure to the regulated proton beam was reduced. Exposure time is an important factor not only in minimizing radiation damage to the sample (several megarads per measurement) but also in preventing loss of data due to random transients that fre-

quently occur in the regulated beam. Any fluctuation in the 0.1% power regulation during a measurement results in a loss of that measurement. Decreasing the exposure time by a factor of about 5 has greatly increased the reliability of the overall system. With the present system, few data are lost due to accelerator transients, resolution on the heat defect-damage curve is improved and statistical fluctuation of the data has been reduced.

Final measurements have now been completed on linear polyethylene, polystyrene plus 20% carbon, and graphite. Work is in progress for measurement of the heat defect in LEXAN plastic. Measurement on three

samples of polystyrene give discrepancies which appear to be more than expected due to variation in the plastic composition. Work continues to resolve these differences.

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RADIATION PHYSICS

This research effort is primarily concerned with basic mechanisms through which radiation interacts with matter. Emphasis is on biologically relevant materials. The studies involve the initial interactions by which ionizing particles transfer energy to matter and the subsequent transport and degradation of that energy. The work includes investigation of the partition and distribution of energy in the radiation field and in the irradiated medium, and determination of rates and mechanisms of energy transferred and transported.

Interaction Cross Sections

L. H. Toburen and W. E. Wilson

Investigation of the energy and angular distributions of electrons resulting from proton impact on gaseous targets has continued throughout the past year. Data were obtained for the following target gases: argon, hydrogen, neon, and xenon and final touches were applied to the results for nitrogen started last year. In addition to the results obtained for proton impact, similar measurements were made for incident H_2^+ ions with hydrogen gas targets. To supplement our experimental work, a computer program was obtained from Professor M. E. Rudd, University of Nebraska, which enables us to calculate the energy distribution of ejected electrons and compare these results with our measurements. This calculation is based on binary encounter theory of the Gerjuoy-Vriens⁽¹⁾ type and, although semiclassical in nature, it has been shown⁽²⁾ to be a special approximation of the plane-wave Born approximation. The agreement between the measured and calculated electron

energy distributions has been quite good: within a factor of 2 throughout most of the ejected electron energy spectrum. The electron energy distributions discussed above are differential in electron energy only; experimental values are obtained from our measured doubly-differential cross sections by an integration with respect to emission angle. There is very little work available for comparison to our measured doubly-differential cross sections and where calculations do exist, such as for protons on hydrogen, the agreement is not at all satisfactory. Total ionization cross sections which are obtained by integrating our measured cross sections, with respect to both electron emission angle and energy, agree quite well with previously published values obtained by other experimental techniques. Likewise, a deduction of the molecular hydrogen ion breakup cross section from our measurements of H^+ and H_2^+ impact on H_2 is in good agreement with previous values obtained by an entirely different method.

Our study of electron emission cross sections is now being directed toward the effects of molecular bonding on electron energy and angular distributions and how these effects may influence the transfer of energy from a proton track.

Energy Partition and Stopping Power

W. E. Wilson

Doubly-differential electron ejection cross sections for fast protons in gases were analyzed to give the total ionization cross section and mean kinetic energy of first generation secondary electrons (FGSE) in an ionizing event. The results provide the "partial" stopping powers for ionization and kinetic energy of FGSE which have been compared with the total stopping power for fast protons in hydrogen gas.

For fast protons in molecular hydrogen gas, the fraction of the total stopping power which goes to overcome the binding energy of the ionization electron is about 0.33 for 100 keV protons and decreases monotonically to approximately 0.21 at 2 MeV. The fraction of the stopping power which is carried away as kinetic energy by FGSE increases from 0.56 to about 0.65 over the same proton energy range.

The remaining fraction of the total stopping power is consistent with theoretical estimates of excitation cross sections of the residual ions and atoms.

A further breakdown of the partial stopping power for FGSE indicates that about 10% goes into producing subionization secondaries (i.e., with kinetic energy less than the ionization potential of the gas), 40% goes into electrons with energy less than 100 or 200 eV and the remaining 50% is carried away by FGSE with energies greater than 100 or 200 eV.

This information gives, for the first time, a quantitative picture of the energy partition of energetic protons at the beginning of the physical stage of radiation action.

Time Resolved Emission Spectroscopy

M. L. West

Single photon counting techniques have been used to study the fluorescence response and time dependence of organic liquids excited by ionizing and nonionizing radiation. Differences obtained by these modes of excitation give time-correlated information about events immediately (10^{-9} sec) following deposition of the ionizing radiation. In the past, differences in decay and emission spectra observed by several experimenters, each using one of several modes of excitation, have raised questions of sample purity and experimental conditions as possible causes of these differences. Since we use the same sample preparation and measurement techniques with all modes of excitation, problems associated with sample integrity are

greatly simplified. High purity samples were prepared and used in these fluorescence studies. The various purging techniques--N₂ bubbling, He bubbling, and freeze-thaw-pump--have been compared and found to give identical results under controlled experimental conditions.

Precise decay curves were measured for proton and ultraviolet excitation of dilute (0.01M) solutions of benzene in cyclohexane over the temperature range 10 to 50 °C. Fluorescence decay under ultraviolet excitation is exponential and can be adequately described by a decay rate of the form

$$k = k_0 + k_1 e^{-w/Kt}$$

where k_0 is a temperature independent rate and k_1 and w are the frequency factor and activation energy of internal conversion quenching. Fluorescence decay under proton excitation was found to be nonexponential and suggests a diffusional, possibly bimolecular, quenching model whereby excited benzene molecules created along a localized proton track are quenched as they diffuse from the initial reaction site. Quenching by similar excited molecules or by transient species created within the track are the two likely candidates. We are attempting to build a simple model based on diffusion about a single proton track including possible track-track interactions that will explain the observed decay curves over a wide range of beam current (dose rate), sample concentration, and sample temperature.

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