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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1969
TO THE USAEC
DIVISION OF BIOLOGY AND MEDICINE
VOLUME II: PHYSICAL SCIENCES
PART 2. RADIOLOGICAL SCIENCES
February 1970

AEC RESEARCH &
DEVELOPMENT REPORT

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BNWL-1307
Part 2
UC-48, Biology
and Medicine

PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1969
to the
USAEC Division of Biology and Medicine
VOLUME II: PHYSICAL SCIENCES
Part 2. Radiological Sciences

By

J. M. Nielsen, Manager
and
Staff Members of

Radiological Sciences Department
Environmental and Life Sciences Division

February 1970

BATTELLE MEMORIAL INSTITUTE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

Volume I of this report to the USAEC Division of Biology and Medicine covers work in the life sciences and is issued as **BNWL-1306** in two parts: Biological Sciences, Part I and Ecological Sciences, Part 2. Volume II of this report covers work in the physical sciences and is issued as **BNWL-1307** in three parts: Atmospheric Sciences, Part 1; Radiological Sciences, Part 2; and Instrumentation, Part 3.

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

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. This report, however, represents a significant change in format from the previous reports in that special effort has been exerted to make the report merely 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

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PACIFIC NORTHWEST LABORATORY
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RADIOLOGICAL CHEMISTRY

INHALATION HAZARDS TO URANIUM MINERS

The documented high incidence of lung cancer and respiratory ailments in uranium miners requires the investigation and measurement of the gross composition and particle size of the organic and inorganic aerosols to which the miner is exposed.

Characterization and Procedure
Development for Radon Daughters

R. W. Perkins, J. A. Cooper,
H. G. Rieck, Jr., and P. O. Jackson

The major radiation hazard associated with uranium mining results from pulmonary deposition of radon daughters and subsequent lung cancer development. The possible synergistic effect promoting lung damage by the chemical explosive constituents, diesel fumes, and ore dust particles must be evaluated. A procedure for the continuous direct

measurement of radon daughter concentrations in uranium and in experimental animal chambers as a function of particle size has been developed which employs both gamma-ray and alpha-particle spectrometry for determining air concentrations of RaA (^{218}Po , 3.05 min), RaB (^{214}Pb , 26.8 min), RaC (^{214}Bi , 19.7 min), and RaC' (^{214}Po , 1.6×10^{-4} sec). The measurement of RaC' is provided by both the gamma-ray spectrometric measurement of RaC and the alpha-particle spectrometric measurement of RaC'. Thus, the attenuation of

the RaC' alpha particles is continuously determined and the alpha counting efficiency for RaA is provided. Short successive counting intervals thus provide a continuous measurement of air concentrations. Particle sizes from a maximum to 0.1μ are determined by the cascade impactor stage employed at the air inlet. Subsequent analyses of the filter permit detailed chemical and radiochemical data to be collected for uranium chain members in the aerosol particle range under study. This approach is also feasible for continuous measurement of radon daughter concentrations in the atmosphere. For these measurements, the cascade impactor stage is removed, IPC filter papers are used in place of the membrane filters, and flow rates of approximately 50 scfm are employed.

A procedure was developed for the determination of ^{226}Ra in animal tissues utilizing a scintillation measurement of ^{222}Rn plus its alpha-emitting daughters, ^{218}Po and ^{214}Po . The purified radium solution is sealed in a counting vial with a mixture of mineral oil, ZnS scintillator, and 1 mm glass beads. The spherical shape of the beads keeps the radium solution from contacting the scintillator. The background for a vial containing scintillator but no radium solution is <0.2 count/min, with an efficiency of 0.32 count/disintegration based on ^{222}Rn plus ^{218}Po and ^{214}Po . The

radium separation scheme modifies a BaSO_4 coprecipitation technique by adding a metathesis step to convert the BaSO_4 to BaCO_3 .

Elemental Deposition in Lung Tissues

L. A. Rancitelli, R. W. Perkins,
and A. D. Renzetti, Jr.*

Samples of lung tissue from individuals with no history of mining experience are being analyzed by a technique of instrumental neutron activation analysis. The concentrations of most elements in the lungs of normal individuals are rather constant as evidenced by the concentrations measured in six samples from one individual. However, individuals with chronic occupational exposures exhibit increased deposition of specific elements compared to normal individuals. The lung tissue from an individual with a history of heavy element work experience contained 0.8 ppm thorium, compared to <0.1 ppm in normal subjects. The lung tissue from a uranium miner contained 2.1 ppm uranium and lung tissue from an individual with uranium processing experience contained 27 ppm uranium, while normal individuals were found to have concentrations below 1 ppb.

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

Neg 0695811-13

Neg 0671020-1

	Ag	Br	Co	Cr	Cs	Fe	Hg	K	Na	Rb	Sb	Sc	Se	Zn
LEFT LUNG														
UPPER LOBE	0.023	66	0.029	1.1	0.027	510	0.54	11,900	8,400	17	0.048	0.0041	0.95	67
LINGULA	0.024	64	0.029	0.9	0.026	430	0.60	11,600	7,400	19	0.037	0.0034	1.0	64
LOWER LOBE	0.087	62	0.028	1.1	0.028	450	0.59	10,200	7,600	24	0.046	0.0039	0.95	65
RIGHT LUNG														
UPPER LOBE	0.005	48	0.026	0.7	0.017	340	0.45	11,500	5,000	19	0.026	0.0023	0.98	49
MIDDLE LOBE	0.017	58	0.025	0.8	0.027	390	0.30	10,300	6,200	20	0.026	0.0028	0.89	61
LOWER LOBE	0.003	57	0.021	0.8	0.022	330	0.36	9,000	6,000	16	0.030	0.0027	0.68	68
AVERAGE	0.026	59	0.026	0.9	0.024	410	0.47	10,800	6,800	19	0.036	0.0032	0.91	62
±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
σ	0.031	6	0.003	0.2	0.004	70	0.12	1,100	1,600	3	0.010	0.0007	0.12	7

Neg 0694079-2

Trace Element Analysis of Human Lung Tissue (μg/g Dry Tissue)

PUBLICATIONS

L. A. Rancitelli, J. A. Cooper, and R. W. Perkins. "The Multielement Analysis of Biological Material by Neutron Activation and Direct Instrumental Techniques," Proceedings of the 1968 International Conference: Modern Trends in Activation Analysis, NBS Special Publication 312, vol. 1, pp. 101-

109. Gaithersburg, Maryland, October 7-11, 1968.

L. A. Rancitelli, T. M. Tanner, and W. A. Haller. "A High Purity Cellulose as a Possible Biological Reference Material," Proceedings of the 1968 International Conference: Modern Trends in Activation Analysis, NBS Special Publication 322, vol. 11, pp. 1306-1310, Gaithersburg, Maryland, October 7-11, 1968.

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.

Radionuclide Distribution in the
Alaskan Ecosystem

C. E. Jenkins

Some 16 radionuclides are being traced through a simple food web of the ecosystem to identify pathways, retention, and concentration factors on three successive trophic levels, lichen-caribou-wolf or man. Each successive trophic food level discriminates against ^{54}Mn , ^{60}Co , ^{88}Y , ^{95}Zr - ^{95}Nb , ^{103}Ru , ^{106}Ru , ^{125}Sb , ^{144}Ce , ^{155}Eu , ^{210}Pb , and ^{214}Bi , whereas ^{22}Na , ^{65}Zn , $^{110\text{m}}\text{Ag}$, and ^{137}Cs are concentrated on each level. Radium-226 is highly concentrated by caribou but not by wolf in the bone tissue. Many of the discriminated radionuclides, as well as radium, are bone seekers and are effectively removed from the third trophic level by their deposition in bone tissue. Sodium-22 is concentrated on all trophic levels by approximately tenfold for caribou-lichen and twofold for wolf-caribou, whereas ^{137}Cs varies from one- to twofold for caribou-lichen and two- to threefold for wolf-caribou. Radionuclide accumula-

tions in lichen vary by species and location; caribou and wolf accumulations are age, sex, and season dependent.

The 16 radionuclides, as well as ^{55}Fe , are being measured in five salmon species from Kotzebue, Alaska, to Sacramento, California. The concentrations of ^{55}Fe and ^{54}Mn decrease tenfold from northern to southern waters. Iron-55 concentrations and specific activities are uniform west of Canada, suggesting that the major salmon feeding grounds are near the Aleutian Island chain. King and Coho salmon from more southern waters exhibit two different ^{55}Fe concentration patterns, which indicate that their migratory patterns are more complex than those of Alaskan salmon. Cesium-137 and cobalt-60 concentrations are fairly uniform throughout all regions, although ^{137}Cs does have a maximum at about 46 degrees N. Zinc-65 concentrations have been observed in salmon samples from all regions. Peak ^{65}Zn concentrations are observed for immature salmon obtained north of the Columbia River. These concentrations decrease rapidly south

of the Columbia River. Zinc-65 concentrations in northern Alaskan salmon are considerably less than those for southern California.

Trace Element Concentrations in Pacific Salmon

L. A. Rancitelli

Since the biological uptake rates of fallout radionuclides are related to the availability of their stable element counterparts within the environment, an extensive study is being made of the trace element concentrations in several principal food sources of the Alaskan locale. Samples of muscle and liver from five species of Pacific salmon have been analyzed by a multielement technique of neutron activation analysis. The concentrations of 14 elements--Na, K, Cs,

Rb, Hg, Cr, Fe, Zn, Co, Sc, Sb, Se, Ag, and Br--have been characterized in the salmon by species and sex. Rubidium and potassium acted very much alike, while cesium was preferentially concentrated in the organism. Similar information has been accumulated for terrestrial food sources such as caribou.

PUBLICATIONS

C. E. Jenkins. *Radionuclide Distribution in Bering Sea and Pacific Ocean Salmon*, BNWL-SA-3023. Battelle-Northwest, Richland, Washington. To be published in *Health Physics*, 1970.

C. E. Jenkins. "Radionuclide Distribution in Pacific Salmon," *Health Physics*, vol. 27, pp. 507-512. 1969.

	Relative Values	
	Rb/K ($\times 10^4$)	Cs/K ($\times 10^6$)
Chum Salmon (9)*		
Muscle, Dorsal	2.4 $\pm$ 0.7	4.2 $\pm$ 0.3
Liver	4.1 $\pm$ 1.5	3.7 $\pm$ 1.0
King Salmon (8)		
Muscle, Dorsal	2.5 $\pm$ 0.3	10.0 $\pm$ 2.2
Liver	3.3 $\pm$ 0.4	5.7 $\pm$ 1.6
Pink Salmon (4)		
Muscle, Dorsal	2.2 $\pm$ 0.2	7.2 $\pm$ 3.2
Liver	3.6 $\pm$ 0.4	4.7 $\pm$ 1.2
Red Salmon (4)		
Muscle, Dorsal	2.1 $\pm$ 0.3	5.3 $\pm$ 0.7
Liver	2.8 $\pm$ 0.9	1.7 $\pm$ 0.4
Silver Salmon (4)		
Muscle, Dorsal	2.7 $\pm$ 0.5	6.7 $\pm$ 1.3
Liver	3.6 $\pm$ 0.5	3.5 $\pm$ 1.6
Sea Water	3.2	0.7

* Number of individuals sampled.

Neg 0695694-1

Neg 0695694-2

Frequency Distribution of Zinc in Pacific Salmon. Concentration (dry basis) in liver (left graph) and in dorsal muscle (right graph).

Neg 0695811-7

Concentrations of ^{137}Cs and ^{22}Na in Muscle Tissue of Caribou, Wolf, and Lichen

PHYSICAL AND RADIOLOGICAL CHEMISTRY OF OCEAN SOLUTIONS

The rates and mechanisms of the chemical and mixing processes in the ocean are being investigated by use of the ocean's natural elemental anti radionuclide tracers. These studies as a function of depth over wide areas of the ocean are allowing the determination of factors governing vertical diffusion and the determination of oceanographic pollutant concentrations and their pathways to man.

The Distributions of Mercury, Cobalt, and Gold in Ocean Water

D. E. Robertson and E. D. Wood*

The distribution of mercury as measured by neutron activation analysis in Atlantic Ocean water and its adjacent seas averaged 0.10 μg Hg/liter with a range from 0.06 to 0.19 μg /liter. Pacific Ocean water contained significantly higher levels, averaging 0.30 μg /liter. Columbia River estuary samples of low salinities averaged 0.11 μg /liter, indicating a dilution of mercury in estuarine waters by the entering river. The chemical and biological processes operating in the ocean cannot account for these high mercury values; thus, they may be pollution from terrestrial sources.

In oceanic and coastal waters, cobalt distributions are extremely variable, with the average concentrations in specific regions of the world's open ocean varying only two- to threefold. Coastal waters normally contain higher cobalt levels, especially near river estuaries, where relatively high concentrations

are introduced from rivers. The highly variable distribution of cobalt over relatively small areas of the ocean is perplexing as no known processes appear to be sufficiently great to create the observed variations. Assimilation of cobalt by colonies of phytoplankton and marine organisms cannot account for these variations. Other processes which might alter the cobalt distribution, such as chemical precipitation, submarine volcanization, adsorption onto detritus and mineral constituents, and contaminated effluents discharged from ships, do not appear to be major factors.

The geochemistry of gold in the marine environment is being investigated by studies of ionic species and suspended particulate matter collected on filters. Gold concentrations in suspended particulate matter ranged from 10^{-11} g/liter to 10^{-8} g/liter, with the average being 5×10^{-10} g/liter of seawater; ionic gold was approximately 10^{-9} g/liter. Preliminary results indicate that a significant fraction of the total gold in seawater may be neither particulate nor ionic.

* AEC Graduate Fellow

Radionuclide Distribution in the
World's Oceans

W. B. Silker

Latitudinal influence on the deposition of cosmogenic ^7Be and fission products has been measured in surface seawater samples collected in the North Pacific Ocean off the coasts of Central and North America.

Beryllium-7 concentrations decreased from 600 to 700 dis/min/m³ in equatorial waters to a minimum of 20 to 30 dis/min/m³ between 20 degrees N and 25 degrees N and then increased to approximately 400 dis/min/m³ at 45 degrees N.

Vertical diffusion coefficients across the main thermocline in the open ocean between Hawaii and California ranged from 0.3 to 0.6 cm²/sec, as determined from vertical concentration gradients of ^7Be . Apparent vertical diffusion coefficients of 3.6 and 7.1 cm²/sec have been obtained from samples collected from Oregon coastal waters. These high values probably reflect advective mixing processes characteristic of this location. In general, the short-lived radionuclides are uniformly distributed throughout the mixed layer of the ocean and decrease significantly in concentration in the water mass below the top of the main thermocline.

Equipment and techniques utilizing a filtration sorption bed and 1600 meters of 2.5 cm ID pipe have been

used to collect 14 radionuclides from 7000 liters of ocean water in a few hours. The system also allows the collection of uncontaminated 1-liter samples from the surface to 1600 meters for trace element analyses.

PUBLICATIONS

D. E. Robertson. The Distribution of Cobalt in Oceanic Waters, BNWL-SA-2796. Battelle-Northwest, Richland, Washington. To be published in Geochim. Cosmochim. Acta, 1970.

D. E. Robertson. The Mercury Content of Oceanic Waters, BNWL-SA-2968. Battelle-Northwest, Richland, Washington. To be published in Science, 1970.

W. B. Silker, R. W. Perkins, and H. G. Rieck, Jr. The Battelle Large Volume Water Sampler, BNWL-SA-3016. Battelle-Northwest, Richland, Washington. To be published in Deep-Sea Research, 1970.

PRESENTATIONS

R. W. Perkins, D. E. Robertson, and L. A. Rancitelli. "Nuclear Methods for the Analysis of Radionuclides and Trace Elements in Seawater, Organisms, and Sediments," IEEE Nuclear Science Symposium, San Francisco, California, October 29-31, 1969.

D. E. Robertson, L. A. Rancitelli, and R. W. Perkins. "Multielement-Analysis of Seawater, Marine Organisms, and Sediments by Neutron Activation Without Chemical Separations," Gordon Research Conference on Chemical Oceanography, Meriden, New Hampshire, July 14-18, 1969.

Neg 0695811-9

Regional Average Concentrations of
Mercury and Cobalt in Oceanic and
Coastal Waters

Neg 0682574-cn

Neg 0695621-1

Neg 0692903-7cn

GEOCHEMICAL OCEAN SECTION STUDY (GEOSECS)

The primary objective of the program is to systematically survey the chemical and radiochemical components of the world's oceans, including overlying air and underlying sediments.

Optimum Sampling and Storage Techniques for the Trace Element Analysis of Seawater - Preliminary Results of the September, 1969, Pre-GEOSECS Interlaboratory Calibration Cruise
D. E. Robertson

This investigation develops the necessary information to describe oceanic mixing processes, organic productivity, air-sea interactions, and trace element and radionuclide distributions. Detailed measurements of the oceanic constituents in water profiles taken along Arctic to Antarctic sections of the oceans will provide extensive physical and chemical data on the same set of samples.

In September, 1969, a pre-GEOSECS intercalibration and testing cruise was conducted off San Diego, California, aboard the Scripps Institution of Oceanography R/V "Thomas Washington." During this cruise, seawater samples were collected at 16 depths between the surface and 4000 meters in 30-liter modified Niskin bottles for interlaboratory comparison of the trace element content of the water. Each 30-liter sample was divided into four

aliquots and treated by (1) filtering through a Nucleopore filter followed by addition of HCl, (2) filtering but with no acidification, (3) no filtering but with acidification, and (4) no filtering and no acidification. The samples were submitted to Battelle-Northwest, Woods Hole Oceanographic Institution, and Yale University for analysis in cleaned polyethylene bottles. These samples will provide the information needed to determine the optimum sampling and storage methods to be used during the GEOSECS expedition, and to determine each laboratory's capability for measuring various trace elements.

Analysis at Battelle-Northwest consists of neutron activation and direct gamma-ray spectrometry utilizing multidimensional and Ge(Li) analyzers. This purely instrumental method permits the measurement of Sr, Rb, Cs, U, Sb, Co, Sc, Fe, Zn, and Hg in a single 25-ml sample of seawater. In general, the trace element concentrations of waters sampled during the pre-GEOSECS

cruise were typical of those previously found in other oceanic waters. Uranium, cesium, and antimony showed very little variation with depth. Cobalt concentrations were typically low but mercury levels significantly higher than in Atlantic waters,

which averaged approximately 0.1 μg Hg/liter. The relatively high mercury levels found in the pre-GEOSECS samples were consistent with previous measurements of Pacific Ocean waters, which were also high in mercury.

The Effect of Storage Procedures on the Trace Element Analysis of Seawater Samples Collected During the 1969 Pre-GEOSECS Interlaboratory Calibration Cruise off San Diego

Depth (m)	Storage Procedure*	Concentration micrograms/liter							
		Fe	Zn	U	Cs	Hg	Sb	Co	Se
10	NF - NA	3.1	0.87	3.4	0.30	0.24	0.20	0.015	0.0006
10	F - NA	3.2	0.71	3.4	0.29	0.28	0.20	0.017	0.0005
10	F - A	3.1	1.4	3.5	0.29	0.51	0.19	0.034	0.0004
450	NF - A			3.5	0.30		0.21	0.048	0.0005
450	F - A			3.5	0.31		0.27	0.047	0.0005
650	NF - NA			3.5	0.32		0.22	0.022	0.0006
650	NF - A	2.7	0.87	3.8	0.32	0.61	0.26	0.021	0.0005
650	F - NA			3.7	0.32		0.25	0.025	0.0005
650	F - A			3.6	0.31		0.25	0.024	0.0005

* F - A - Filtering through a Nucleopore filter followed by addition of HCl

F - NA - Filtering but with no acidification

NF - A - No filtering but with acidification

NF - NA - No filtering and no acidification

Neg 0671870-2

The Increased Resolution of Ge(Li) Spectrometers Allows
the Determination of Multiple Trace Elements in Seawater
Following Neutron Activation

IN- CLOUD SCAVENGING

The identification of cloud processes which are instrumental in transferring and removing airborne contaminants into or from the precipitation element; requires studies of the in-cloud and total wet removal of airborne contaminants.

Cosmogenic Radionuclide Tracers of
In-Cloud Scavenging Processes

R. W. Perkins, C. W. Thomas,
J. A. Young, and H. G. Rieck, Jr.

Technology has been developed which allows the measurement of the short-lived cosmogenic radionuclides ^{24}Na , ^{38}Cl , and ^{39}Cl in rain during consecutive sampling periods as short as 10 minutes. The technique involves collection of ^{24}Na on cation and ^{38}Cl and ^{39}Cl on anion resin beds, followed by direct counting on multidimensional gamma-ray spectrometers. Measurement precision is better than $\pm 10\%$, providing excellent resolution for studying in-cloud processes.

Observation of three rainstorms in Washington with this technique suggests the following sequence in rainwater development. Cloud droplets form on condensation nuclei which have been "labeled" with the cosmogenic radionuclides. During subsequent growth of the cloud droplets through coalescence and condensation, additional collection of newly formed cosmogenic radionuclides appears small or negligible. During

their in-cloud development, the raindrops may be subjected to several cycles of partial evaporation followed by further coalescence and condensation, particularly in light rains.

Light rains have spent a substantially longer period in their development than heavy rains. They show both higher radionuclide concentrations and higher long- to short-lived radionuclide ratios. The time period for raindrop development ranged from approximately 30 to 130 minutes in the light to medium rains which were studied, based on the $^{39}\text{Cl} : ^{38}\text{Cl}$ ratio in rainwater relative to that of the atmosphere. Rainout coefficients for the storms were in the range of 10^{-3} to 10^{-2} sec^{-1} but could be some two- to threefold lower, depending on the effective altitude of the cloud top. These rainout coefficients were determined by comparing the radionuclide deposition per unit area at ground level (decay corrected to the time of nucleation) to the radionuclide content per unit area of cloud in an air column of cloud thickness. The

apparent "time since previous rain-out" in the rainstorms ranged from 30 to 200 minutes from estimates of the ^{24}Na : ^{38}Cl ratio in the rainwater. These very short values would require that essentially complete rainout of

the cosmogenic radionuclides occur at these time intervals. This apparent behavior is in agreement with the observed high rainout coefficients.

Neg 0695811-5

Rainout Coefficients for ^{38}Cl Observed During Two Rainstorms, o Denotes 2-15-69 Reading and Δ Denotes 4-3-69 Reading

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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.

Multielement Analysis of Waters by Neutron Activation

T. M. Tanner and L. A. Rancitelli

A procedure has been developed which eliminates ^{24}Na interference in neutron activation procedures and separates a radionuclide mixture into two groups for analysis. Separations are accomplished by passing the sample through anion exchange resins in the hydroxide-sulfide-carbonate form, allowing alkali metal ions and alkaline earth ions to pass through the column.

Further separations from sodium are accomplished by precipitating K, Rb, Cs, and the alkaline earths with sodium tetraphenyl boron. The result of this procedure is three fractions --the anion exchange resins, the tetraphenyl boron precipitate, and the separated sodium. This procedure, which allows the determination of concentrations of 10 to 20 elements in a single sample, has been used for the analysis of Columbia River water, rainwater from the Olympia National Park, and ice cores

from Greenland. The method is being applied to studies of precipitation scavenging mechanisms in the atmosphere, environmental pollution, and elemental ecological transport.

Lead Analysis Employing Fast Neutron Activation

L. A. Rancitelli, T. M. Tanner, and J. C. Langford

The advent of the Fast Flux Test Facility offers a new approach to the measurement of many specific elements by fast neutron activation. An example is the measurement of lead by its isotope ^{204}Pb , which has an appreciable inelastic scattering cross section for the production of $^{204\text{m}}\text{Pb}$ ($t_{1/2} = 67$ min). Lead foils have been bombarded with 3 MeV neutrons to produce $^{204\text{m}}\text{Pb}$, which was subsequently used to develop rapid, high-yield radiochemical separations of lead from biological tissues. A few parts of lead per billion can be measured in biological tissue assuming a neutron flux of 10^{16} neutrons/cm²/sec.

MULTIELEMENT ANALYSIS OF TRACE CONSTITUENTS IN NATURAL WATER
BY NEUTRON ACTIVATION, TWO-GROUP CHEMICAL SEPARATIONS AND
GAMMA-RAY ANALYSIS

Trace Element Concentrations of Rain-
water Samples Taken a Few Minutes
Apart at Olympic National Park,
Washington

(parts per billion)

Sample No. 1 Sample No. 2

Ag	1.0	1.1
As	1.3	1.4
Au	<0.02	<0.02
Br	3.8	1.6
Cl	1400	690
Co	0.045	0.042
Cr	<2	<2
Cs	0.07	0.07
Cu	3.4	2.8
Fe	35	<20
Hg	<1	<1
K	630	430
Mh	2.9	2.3
Na	760	380
Rb	<2	<2
Sb	0.18	0.14
Sc	0.003	<0.001
Se	0.07	0.07
Zn	19	20

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RADIOACTIVE FALLOUT RATES AND MECHANISMS

Studies of the meteorological, chemical, and radio-chemical compositions of atmospheric aerosols from ground level to maximum attainable altitudes are being made to determine the rates and mechanisms by which airborne radionuclides become attached to the aerosol, the origin and chemistry of the host aerosol, and the rates and mechanisms of mixing and scavenging processes in the atmosphere.

Cosmogenic Radionuclides in the Environment

J. A. Young, C. W. Thomas,
N. A. Wogman, R. W. Perkins, and
H. G. Rieck, Jr.

The disintegration rates of the short-lived cosmogenic radionuclides ^{39}Cl (55 min), ^{38}Cl (37 min), and ^{24}Na (15 hr) and some longer-lived radionuclides have been measured at altitudes from ground level through 19 km for use as tracers of precipitation and short-term atmospheric mixing processes. The production rates of ^{39}Cl , ^{38}Cl , ^{38}S , $^{34\text{m}}\text{Cl}$, ^{28}Mg , ^{24}Na , and ^{18}F produced by spallation of ^{40}Ar with cosmic rays at 18 km have also been determined for the first time with equilibrium concentrations of 483, 320, 35.1, 14, 5.35, 20.4, and 3.7 dis/min/ 10^4 S.C.F. of air respectively. In addition, large crystal multidimensional gamma-ray spectrometers have been used to investigate the aerosol at ground level and to detect the first ^{24}Na and an additional 18 fallout radionuclides in river water following their absorption by ion exchange

resins. Sodium-24 in the river resulted only from dry deposition at ground level since no recent rainstorms had occurred. These data have been used to design an experiment which will measure the rate of dry deposition of radionuclides with half-lives as short as 30 min and to resolve the short-term variations in the deposition rates.

Atmospheric Trace Elements

L. A. Rancitelli and R. W. Perkins

The atmospheric aerosol, which is a major factor in the airborne transfer of radioactivity, originates from man-made and natural sources. Once injected, these materials can serve as stable tracers of the vertical and horizontal air masses and reflect the movement of attached fallout on cosmic-ray-produced radionuclides.

A technique was developed and applied to measure 19 elements on filters and in rainwater to establish the gradient of several trace

elements from the troposphere through the lower stratosphere. The concentrations of Ag, Co, Cr, Fe, Sb, Sc, and Zn which were measured by instrumental neutron activation analysis in the atmosphere over a two-year period varied from 10- to 100-fold over the altitude range of 3 to 15 km. Concentrations at a given altitude varied severalfold as a function of time. A comparison of the relative concentrations observed in the atmosphere with those in metropolitan aerosols, the earth's crust, seawater, and rainwater collected at points remote from possible sources of pollution indicates that while atmospheric Fe, Co, and Sc concentrations are compatible with the injection of material from the earth's crust, the concentrations of Ag, Cr, Sb, and Zn in the atmosphere are too high to have originated from natural sources.

The observed concentration of scandium at 15 km is approximately 10^{-18} g per gram of air. Although significant pollution from industrial sources has been observed in the stratosphere, the concentrations of many of the trace elements are in approximately terrestrial ratios. As little as 0.5 kg of elements such as rubidium and iridium could increase the concentration in a 13.7 to 16.8 km atmospheric shell by tenfold. Since higher shells of the atmosphere contain lower concentrations of the trace elements, comparable or smaller amounts of tracer could be used to tag these atmospheric sections.

Preliminary Results from Radio-nuclide Measurements Made During BOMEX

J. A. Young, H. G. Rieck, Jr.,
D. E. Robertson, and W. B. Silker

During the experimental phase of the Barbados Oceanographic and Meteorological Experiment, the concentrations of ^7Be and several nuclear-weapons-produced radionuclides were measured in air, seawater, and rain samples. Because of its relatively short half-life, ^7Be is confined primarily to the top 50 meters of the ocean. Observed changes were not induced by rain since the largest amount of ^7Be contributed by any one rainfall was less than 1% of the total amount in the sea. During the experimental periods, the rate of addition of ^7Be to the sea by rainfall was only 10% to 15% of its rate of decay in the sea. It does not seem likely that turbulent transport across the sea surface could cause rapid changes in concentrations which generally occurred simultaneously throughout the top 30 meters. The decreases in concentration throughout some vertical profiles could not have been caused by vertical transport; therefore, the changes in concentration were probably caused by the horizontal transport of water of different activities through the array. Fresh water was present during the second two weeks of June. The high activities observed during this period were probably caused by Amazon Basin water

since it is produced primarily by rain, with concentrations averaging approximately 1000-fold that of seawater.

Although the activities of ^7Be and the nuclear-weapons-produced radionuclides in the atmosphere rapidly increased above 9.1 km, they remained fairly constant from 0.3 to 9.1 km. Rather large changes in concentrations with time and location were observed. On the average there was a minimum in the activities of all of the radionuclides at 1.5 km, probably caused by rainout. At a specific altitude, some vertical profiles showed activity minimums for the bomb-produced radionuclides but not for ^7Be . These minimums probably resulted from intrusions of southern hemisphere air since the activities of the bomb-produced radionuclides are significantly lower in the southern hemisphere than in the northern, while the ^7Be activities are about the same. Some profiles showed large activity minimums at 9 km which were not caused solely by intrusions of southern hemisphere air since ^7Be showed the same activity minimum as the bomb-produced radionuclides. These minimums were probably the results of rainout. Some of the air samples collected near the sea surface showed increasing radionuclide activities from 0.3 to 21 meters.

Atmospheric Fallout

C. W. Thomas and H. G. Rieck, Jr.

The atmospheric concentrations of 20 radionuclides are being measured at ground level near Point Barrow, Alaska (71 degrees N); Richland, Washington (46 degrees N); Barbados, British West Indies (13 degrees N); and Rio de Janeiro, Brazil (23 degrees S). In the northern hemisphere, nearly all concentrations are equal to or greater than those measured in the previous two years and reflect the high yield thermonuclear testing by the Peoples Republic of China. The concentration increase was greatest at the mid-latitudes. The shape of the annual spring maximum at Point Barrow, Alaska, was notably different, showing a second peak in July because of the Chinese thermonuclear tests of previous years. The concentrations of the medium-lived fission products ^{95}Zr , ^{95}Nb , ^{103}Ru , and ^{141}Ce achieved 1963 concentration values.

Air filter samples collected at ground level during the period 1963 through 1968 at Richland, Washington, have been analyzed for ^{90}Sr and ^{55}Fe concentrations. The air concentration of ^{90}Sr showed a seasonal variation comparable to that of ^{137}Cs . The resident half-time of ^{90}Sr measured from the 1963 to 1967 concentrations was approximately 11 months. Seasonal variations in

^{55}Fe concentrations are created by stratosphere-troposphere exchange and plant operations.

The cosmogenic radionuclide ^{22}Na attained the same concentration in the 1969 spring maximum that was measured in the spring maximums for 1967 and 1968. This indicates that the large reservoir of ^{22}Na produced in the nuclear tests of 1961 and 1962 has been depleted and measurements now reflect cosmogenic ^{22}Na . Beryllium-7 atmospheric concentrations continue to show the characteristic spring maximums and fall minimum, providing a base line for air-sea transport studies at various latitudes.

Neg 0692929-3

^7Be Concentrations in Sea Water at
15 Degrees N, 57 Degrees W

Radionuclides in the Plowshare

Event Schooner

W. B. Silker

Radionuclides present in the high cloud from the Plowshare event Schooner have been measured in air, in deposition samples collected 100 and 250 miles from ground zero, and in the base surge cloud from stations at Spokane, Washington, and Great Falls, Montana. The major radioactivities were associated with $>3 \mu$ particles; however, their relative distributions on smaller particles were similar to those observed in previous events. The relative concentrations of tungsten and molybdenum radioisotopes were enriched two-fold in air samples taken at the three far out locations when compared with those less than 100 miles from ground zero.

Neg 0695811-6

^7Be as a Function of Time in Sea
Water

Neg 0695811-10

^7Be and ^{95}Zr Disintegration Rates at 18 Degrees N,
55 Degrees W on 5-9-69

Neg 0695811-8

^7Be and ^{95}Zr Disintegration Rates at 18 Degrees N,
55 Degrees W on 5-9-69

Neg 0695811-1

Concentrations of ^{137}Cs in Ground-level Air. Solid curve represents data collected at Richland, Washington (46 degrees N) and dotted line represents data obtained near New York City (41 degrees N)

Neg 0693404-7

Trace Element Concentrations in the Atmosphere versus Altitude; Summation, 35 to 45 Degrees N Latitude, 110 Degrees W Longitude

Neg 0695811-12

Radionuclides from Project Schooner in Order of Increasing Deposition to Air Concentration Ratios

Estimated Concentrations of Rare Elements Between 13.7 and
16.8 km for Predictive Use as Tracers of Atmospheric Aerosols*

Element	Terrestrial Abundances, ppm	Total Weight Element, g/ 10 ¹⁸ ft ³ of Air	Total Weight Element, g/ 5 × 10 ¹⁹ ft ³ of Air	Kilograms of Element Required to Increase Concentration × 10	
		Case 1**	Case 2†	Case 1	Case 2
Sc	5	5,000	300,000	50	3,000
La	18	18,000	900,000	180	9,000
Sm	6.5	7,000	400,000	70	4,000
Gd	6.4	6,000	300,000	60	3,000
Pr	5.5	6,000	300,000	60	3,000
As	5	5,000	300,000	50	3,000
Hf	4.5	5,000	300,000	50	3,000
Dy	4.5	5,000	300,000	50	3,000
Yb	2.7	3,000	200,000	30	2,000
Er	2.5	3,000	200,000	30	2,000
HO	1.2	1,000	50,000	10	500
Eu	1.1	1,000	50,000	10	500
Tb	0.9	900	50,000	9	500
Lu	0.8	800	40,000	8	400
Tm	0.2	200	10,000	2	100
In	0.1	100	5,000	1	50
Ag	0.1	100	5,000	1	50
Se	0.09	90	5,000	0.9	50
Au	0.005	5	300	0.3	3
Te	0.002	2	100	0.1	1
Ru	0.001	1	50	0.05	0.5
Ir	0.001	1	50	0.05	0.5

* Using a value of 10^{18} g of Sc/g of air and assuming an average air density of 0.2 g/liter. All other elements are ratioed to Sc using terrestrial abundances.

** A 2 degree latitude band at the equator from 13.75 to 16.8 km

† Entire earth's atmosphere between 13.75 and 16.8 km

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IRON-55 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, some of the same samples are used to measure Pb, ^{210}Pb , and ^{210}Po in the biosphere.

Iron-55 in Man and Animals

J. C. Langford and C. E. Jenkins

The ^{55}Fe and Fe concentrations measured in men and women's blood samples from Alaska, Brazil, Colombia, France, Gabon, Italy, Nigeria, Saudi Arabia, Spain, and Richland, Washington, U.S.A., correlate well with ^{55}Fe levels in the atmosphere. The concentration varies as rainfall rate at the collection site. Concentrations of ^{55}Fe in cattle blood correlate with the expected concentration in vegetation and follow a biological half-life of 1.72 years. Iron-55 in Richland, Washington, residents is presently decreasing at 7 nCi per year and is 2.2 times the level in cattle. Iron-55 in Alaskan fish-eating natives is tenfold higher than in Richland residents and the level is declining at a more rapid rate, reflecting the much higher turnover rate for iron in the marine biosphere.

Iron-55 concentrations are being measured currently in all five species of North American salmon at various Pacific locations. These measurements permit the distribution of ^{55}Fe to be studied as a function of latitude, salmon species, sex, age, and trophic feeding level. The more northerly feeding salmon are tenfold higher in ^{55}Fe than the southern salmon, with a two- to threefold yearly decline in ^{55}Fe activity. This high rate of turnover is greater than the fallout measured in air and precipitation and suggests that the ^{55}Fe contribution to the marine biosphere is currently coming from the stratosphere. High specific activities in Bering Sea salmon indicate that the stable iron may be low or unavailable to marine organisms in that region. Similar specific activities in different salmon species suggest that they have a common feeding ground.

Particulate Pb, ^{210}Pb , and ^{210}Po in
the Atlantic Ocean

J. C. Langford

Particulate Pb, ^{210}Pb , and ^{210}Po have been measured in Atlantic Ocean surface samples collected between the Bahama Islands and Sierra Leone, Africa. Although the average particulate ^{210}Pb disintegration rate was 20% higher than the particulate ^{210}Po disintegration rate per liter, no trends were observed. The particulate lead concentration decreased to one ninth (2.23 ± 0.02 to $0.25 \pm 0.03 \mu\text{g/liter}$) from 25 degrees 38 min N, 72 degrees 20 min W

to 24 degrees 48 min N, 59 degrees 20 min W.

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Neg 0670677-4

Low-energy X-ray and Gamma-ray Emitters Can be Determined
Directly by the Use of a Thin Window Si(Li) Spectrometer

Neg 0690574-2

A Comparison of the Relative Decrease of ^{55}Fe in Fallout

Neg 0690598-2

Body Burden of ^{55}Fe in Cattle and of ^{55}Fe and ^{137}Cs in Humans from Richland, Washington

RADIATION INSTRUMENTATION - RADIOLOGICAL CHEMISTRY

This program is concerned with the development of radio-chemical 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.

High Efficiency Anticoincidence Shielded Ge(Li) Gamma-Ray Spectro- meters

J. A. Cooper

A Ge(Li) gamma-ray spectrometer has been developed with two 12%-efficiency diodes orientated 180 degrees apart. This orientation doubles the solid angle and improves the sensitivity 1.4-fold over that of a single diode. Both of these diodes are located at the center of a 76 cm diameter by 76 cm thick plastic phosphor anticoincidence shield with a 15 cm diameter by 10 cm deep central cavity to allow space for counting large environmental samples. Both diodes, which are gain and zero stabilized, are operated in anticoincidence with each other and the shield. Resolutions of approximately 3 keV at 1332 keV are obtained even with 10,000-min count intervals. The shield provides an order of magnitude reduction in the Compton interference. The system provides sensitivities for noncoincident gamma-ray-emitting radionuclides of 0.1 dis/min, in addition to a very high sensitivity

for coincident gamma-ray-emitting radionuclides. The sensitivity for a pure source of ^{60}Co is approximately one fifth of that in multi-dimensional gamma-ray spectrometers constructed from 28 cm diameter by 15 cm thick NaI(Tl) crystals.

The parameters most frequently used to describe the quality of a Ge(Li) spectrometer are the resolution, efficiency, peak-to-Compton ratio, and peak-to-total ratio. Although these parameters are related to the sensitivity, their use in defining the overall quality of a spectrometer has often been based on intuition. An analytical expression has been developed which relates the sensitivity to resolution, efficiency, and background interference. This is expressed by $F_2(E_1) = [\theta \times I_2^2(E_1)]^{1/2} / [R(E_1) \times I_{c2}(E_1)]^{1/2}$ where $F_2(E_1)$ is the figure of merit (sensitivity) for the detection of a gamma ray of energy E_1 in the presence of a higher energy gamma ray, θ is the solid angle, $I_2(E_1)$ is the intrinsic efficiency at energy E_1 , $R(E_1)$ is the resolution at energy E_1 , and $I_{c2}(E_1)$ is the intrinsic

Compton efficiency per keV at energy E_1 resulting from gamma-ray two.

The most interesting feature of the equation is that sensitivity can be estimated solely from the geometry of the diode and cryostat. The solid angle is fixed by the cross section and the distance to the source. The resolution is the square root of the sum of the diode resolution squared plus the square of the electronic noise. The diode contribution can be estimated from the gamma-ray energy and the Fano factor; the electronic contribution can be estimated from the diode capacitance, which is a function of the geometry, and the preamplifier noise specifications. The intrinsic efficiencies can be calculated by the Monte Carlo technique. Such calculations give the optimum diode geometry for problems such as the measurement of a peak in the presence of a single higher energy gamma ray or multiple gamma rays.

Performance and Application of a Multiple Coincidence Ge(Li) Gamma-Ray Spectrometer

R. W. Perkins, L. A. Rancitelli, and J. A. Cooper

A new technique, "Multiple Coincidence Ge(Li) Gamma-Ray Spectrometry," provides an improved sensitivity for the measurement of most radionuclides which decay by emission of two or more photons. The detector arrangement requires the use

of a large Ge(Li) diode inside a large NaI(Tl) well crystal. With the sample located on top of the Ge(Li) diode, four 1024 channel spectra are stored simultaneously. The first is an anticoincidence spectrum obtained with the NaI(Tl) crystal acting as an anticoincidence shield, while the other three are spectra in coincidence with NaI(Tl) energy ranges of 0.1 to 0.66, 0.66 to 1.32, and 1.32 to 3 MeV respectively. Feasibility measurements in a similar detector system but using a NaI(Tl) rather than a Ge(Li) diode detector indicate that the anticoincidence spectrum should have a background one fifth to one tenth of that of a normally operated diode; the backgrounds of the three coincidence spectra will range from approximately one third to one thousandth. The Compton interference will be markedly reduced and, in the higher energy coincidence regions, it should be from one tenth to less than one thousandth of that obtained with the best coincidence methods presently in use.

Low-Level Radionuclide Analysis

N. A. Wogman

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An all sodium iodide anticoincidence shielded total absorption spectrometer designed for low-level radionuclide analysis has been fabricated from a 34 cm diameter by 34 cm thick NaI(Tl) ingot which surrounds an 18 cm diameter by

22 cm thick NaI(Tl) principal detector. In addition to the use of ceramic phototubes, which provide a twofold reduction in ^{40}K background, the system construction uses low background materials throughout. The background in the energy range from 0.1 to 3 MeV is reduced from one half to one fifth by the anticoincidence shield. The efficiency of the system for a point source and a 70 ml source is approximately fivefold greater than that of a 7.5 cm diameter by 7.5 cm thick NaI(Tl) crystal. By means of the anticoincidence crystal, the Compton edge for ^{137}Cs is reduced to one sixth to produce a peak to Compton ratio of 160:1. The sensitivity of the system for single gamma-ray emitters is greater than any NaI(Tl) or Ge(Li) multidimensional gamma-ray spectrometer.

Trace Element and Radionuclide Analysis of Environmental Materials

L. A. Rancitelli, N. A. Wogman,
T. M. Tanner, R. W. Perkins,
J. A. Cooper, R. E. Brown*, and
C. Menninga**

The physical properties of a metal or alloy are attributed to the major and minor constituents of the system which can affect its properties or curtail its uses. Sensitive

multiparameter gamma-ray spectrometers employing large-volume NaI(Tl) crystals and normal anticoincidence shielded Ge(Li) detectors have been successfully applied to the measurement of the concentrations of over a dozen elements (several present at levels below 1 ppb) in high purity copper metal and the fracture surface of embrittled and nonembrittled steel following neutron activation. The trace element content of sodium metal has been determined by direct instrumental analysis of the neutron activated trace elements following ion exchange removal of ^{24}Na .

A ^{252}Cf neutron source and a Ge(Li) gamma-ray detector have been used for rapid nondestructive mineral analyses. A one- or two-minute exposure of mineral samples to the thermalized neutron flux from a 0.26 mg ^{252}Cf source, followed by a ten-minute count with an 80 cc Ge(Li) diode, allows the measurement of most elements at concentrations one tenth to one hundredth the levels of economic interest.

Spherical mineral particles of olivine or pyroxene in chondrules from stony meteorites have also been analyzed by neutron activation analysis. Since the chondrules are composed of high temperature minerals inside a fragment with a low temperature history, the major, minor, and trace element compositions provide clues to their origin. The developed method consists of irradiating a sample and flux standards with 6×10^{14} neutrons/cm², followed immediately by Ge(Li) gamma-ray analysis to measure

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the neutron-induced radionuclides ^{24}Na , ^{27}Mg , ^{28}Al , ^{51}Ti , ^{52}V , and ^{56}Mn . The sample is then re-irradiated with the flux standards to an integrated neutron exposure of 1×10^{18} neutrons/cm². The standards and irradiated chondrules are counted after appropriate decay intervals on a Ge(Li) and a multidimensional gamma-ray spectrometer to measure the neutron-induced radionuclides ^{24}Na , ^{42}K , ^{46}Sc , ^{47}Ca , ^{47}Sc , ^{51}Cr , ^{59}Fe , ^{60}Co , ^{65}Zn , ^{86}Rb , $^{110\text{m}}\text{Ag}$, ^{124}Sb , ^{134}Cs , ^{192}Ir , and ^{198}Au . The technique permits the simultaneous measurement of all transition metals except Cu and Ni, the alkali metals, the major elements Ca, Mg, and Al, and the trace constituents Au, Ag, Ir, and Sb.

Meteorites also accumulate cosmogenic radionuclides, which reflect the intensity of the cosmic-ray spectrum, the size of the body, and its breakup history. Using the multidimensional and anticoincidence shielded Ge(Li) gamma-ray spectrometric techniques, the cosmogenic radionuclides ^{22}Na , ^{26}Al , ^{46}Sc , ^{48}V , ^{51}Cr , and ^{54}Mn ; neutron-induced ^{60}Co ; and the primordial radionuclides ^{40}K and ^{208}Tl have been measured in the recently fallen Murchison meteorite. Similar measurements have been made on the Denver meteorite, which fell in 1967.

These instrumental radiochemical techniques have provided major advances in the understanding of the environmental processes.

Neg 0694672-1

Anticoincidence Spectrometer System

Neg 0695811-2

Cutaway View of the 18 cm Diameter Well Crystal Surrounded by its 34 cm Diameter Anticoincidence Shield and 10 cm Lead Cave

Neg 0692948

Gamma-ray Spectrum of Neutron Activated Argentine

Neg 0695811-3

Multiple Coincidence Ge(Li) Gamma-ray Spectrometer Readout
(For Analysis of 22 Common Neutron Activation Products from
Biological and Environmental Samples -
²²Na, ³⁸Cl, ⁴²K, ⁴⁶Sc, ⁵¹Cr, ⁵⁶Mn, ⁵⁹Fe, ⁶⁰Co, ⁶⁴Cu, ⁶⁵Zn,
⁷⁵Se, ⁷⁶As, ⁸²Br, ⁸⁵Sr, ⁸⁶Rb, ^{110m}Ag, ¹²⁴Sb, ¹³⁴Cs, ¹³⁹Ba,
¹⁹⁸Au, ²⁰³Hg, ²³⁹Np)

Neg 0695811-4

Multiple Coincidence Ge(Li) Gamma-ray Spectrometer Readout
for Analysis of 23 Radionuclides in Fallout on Air Filters
and Environmental Materials:
⁷Be, ²²Na, ⁴⁶Sc, ⁵⁴Mn, ⁵⁷Co, ⁵⁹Fe, ⁶⁰Co, ⁶⁵Zn, ⁹⁵Zr, ⁹⁵Nb,
¹⁰³Ru, ¹⁰⁶Ru, ^{110m}Ag, ¹²⁵Sb, ¹³¹I, ¹³⁴Cs, ¹³⁷Cs, ¹⁴⁰Ba
(¹⁴⁰La), ¹⁴¹Ce, ¹⁴⁴Ce(¹⁴⁴Pr), ¹⁴⁷Nd, ¹⁵⁵Eu, ²⁰⁸Tl

Sensitivities for Measurement of Elements in Representative
Minerals by a Rapid ^{252}Cf Irradiation and Direct Counting

Element	Mineral	Radionuclide Daughter(s)	Half-Life	Detectable Concentrations*				
				ppm		percent		
				-1 to 10	10 to 100	0.01 to 0.1	0.1 to 1.0	1.0 to 10
Aluminum	all minerals	^{26}Al	2.30 min					X
Antimony	stibnite	^{122}Sb	2.8 day				X	
		$^{122\text{m}}\text{Sb}$	4.2 min					X
		$^{124\text{m}}\text{Sb}$	21 sec					
Arsenic	arsenopyrite	^{76}As	26.5 hr				X	
Barium	witherite	$^{133\text{m}}\text{Ba}$	38.9 hr					
		^{139}Ba	83 min				X	
Cobalt	cobaltite	$^{60\text{m}}\text{Co}$	10.5 min				X	
Copper	chalcopyrite	^{64}Cu	12.8 hr				X	
		^{66}Cu	5.1 min					
Dysprosium	gadolinite	^{165}Dy	2.35 hr		X			
		$^{165\text{m}}\text{Dy}$	1.26 min					
Fluorine	gadolinite	^{20}F	11 sec			X		
Gold	quartz	^{198}Au	64.8 hr		X			
Indium	chalcopyrite	$^{116\text{m}}\text{In}$	54 min	X				
Magnesium	magnesite	^{27}Mg	9.5 min				X	
Manganese	all minerals	^{56}Mn	2.58 hr			X		
Mercury	cinnabar	^{205}Hg	5.2 min					X
Niobium	columbite	^{94}Nb	6.3 min					X
Molybdenum	molybdenite	^{101}Mo	14.6 min			X		
Scandium	scheelite	$^{46\text{m}}\text{Sc}$	20 sec	X				
Selenium	chalcopyrite	$^{77\text{m}}\text{Se}$	18 sec	X				
Silver	argentite	^{108}Ag	2.4 min		X			
		^{110}Ag	24 sec					
Strontium	witherite	$^{87\text{m}}\text{Sr}$	2.8 hr				X	
Titanium	rutile	^{51}Ti	5.80 min				X	
Tungsten	scheelite	$^{183\text{m}}\text{W}$	5.3 sec				X	
		^{187}W	24 hr					
Vanadium	any mineral	^{52}V	3.77 min		X			

* ^{252}Cf source of 1 mg
Sample weight of 5 kg
Irradiation time of 2 minutes
Counting time of 2 minutes
Ge(Li) diode size of 80 cm^3 and resolution of 3 keV
Expected precision at the stated detectable concentrations is
 ± 3 to 15%.

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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. The studies are restricted primarily to investigation of radionuclides of interest to the nuclear industry which are difficult or impossible to reliably assess in vivo through the use of external radiation detectors such as whole body counters. Because of this restriction, analysis of body excretion is the usual method of diagnosis, quantification of which is accomplished through excretion studies on human test subjects. Since the radionuclides of interest are usually highly radiotoxic, substitute isotopes which have good measurement sensitivity but which are much less radiotoxic are developed for use in human testing studies. Animal studies performed to give basis for human studies also permit correlation of metabolic parameters between man and animals which in turn can give basis to other animal studies.

Plutonium

I. C. Nelson and D. McConnon

Our current interest in plutonium is the development of a diagnostic method to be used in the presence of chelating agents used for therapeutic removal of plutonium. In addition, human testing performed for the above would add basis to the early work of Langham, et al. The process of preparing test quantities of ^{237}Pu , the substitute isotope for several highly toxic plutonium isotopes, involves cyclotron irradiation and, as it turns out, has involved long turn-around times. Two full irradiation sequences were completed this year. The first was an irradiation

of 99+% enriched $^{235}\text{U}_3\text{O}_8$ which resulted in a yield one tenth that of the previously used 93% enriched uranium-235 metal foil experiments but the product was still contaminated with an excessive amount of the long-lived alpha-emitting radionuclides ^{236}Pu and ^{238}Pu . This poorer yield was attributed to non uniformity of irradiation and it was decided to discontinue use of U_3O_8 targets in favor of the more easily prepared metal foil targets. The second experiment consisted of three separate foils irradiated for shorter times at three different energies. This test will provide information relative to the trade

off of ^{237}Pu yield in favor of reduced amounts of contaminating ^{236}Pu and ^{238}Pu . Separation within 24 hr after irradiation was also performed to reduce the ingrowth of these plutonium contaminants from neptunium decay. Analysis of these samples is still in progress.

A plutonium tissue monitor was designed and prototype built which was tested in the morgue of the local hospital. Fabrication of the permanent device is in progress. This device is for use by the pathologist in screening autopsy cases for gross findings of plutonium in liver tissue. Indication of plutonium would alert the pathologist to a need for a more comprehensive tissue sampling program. This work is in support of the national Plutonium Registry.

Promethium-147

D. McConnon and I. C. Nelson

Current efforts are oriented toward a diagnostic method for inhaled promethium-147 oxide. The earlier inhalation experiment utilizing human volunteers and promethium-143 chloride was abandoned on the advice of the RNW Human Testing Committee in favor of inhalation studies performed with the oxide. This advice was based on the fact that the pressing need is for information on the oxide and, although a chloride experiment would be simpler to perform, the justification for human testing was not so clear in the case of the chloride

since it has a much smaller hazard potential. At an acceptable dose level the mass of promethium 143 oxide is inadequate to permit production of the test aerosol. Samarium was the first choice as a carrier for promethium. It is not known if samarium behaves the same as promethium in a biological system. Experiments have been designed to determine this. Initially samarium chloride and promethium chloride will be administered intravenously to a group of rats with subsequent tissue analysis to determine if gross differences exist in the metabolism of samarium and promethium. If no gross differences exist, groups of rats will be exposed to Sm_2O_3 , $^{147}\text{Pm}_2\text{O}_3$, and $(^{143}\text{Pm}, \text{Sm})_2\text{O}_3$ via inhalation to assess the appropriateness of the carrier in the rat. Again, if the results are favorable, similar tests will be conducted on beagle dogs, an animal large enough for whole body examination, to confirm the basis for human testing.

An aerosol generator was designed, fabricated and tested. Some special apparatus was necessary since the equipment is intended for use in human inhalation testing. Particle size studies were also performed to determine particle size distributions as a function of aerosol concentrations and other operating parameters. For the above mentioned samarium carrier and promethium aerosol, experiments will be performed to ensure that the promethium is distributed uniformly throughout the samarium.

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CHARACTERIZATION OF RADIATION-INDUCED FREE RADICAL REACTIONS
IN AQUEOUS SYSTEMS

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 have been assessed by studying their reactivities toward diagnostic reagents, while methods for chemical protection have been tested in synthetic systems resembling living tissue in particular aspects.

Detection of Amino Acid Radicals
in Aqueous Solution

R. N. Diebel and W. D. Felix

Free radicals derived from amino acids have long been postulated to occur in biological fluids but elaborate techniques are required for their detection and structural determination in fluid media where their existence is quite transient.

Radicals derived from hydroxyl-radical attack on glycine and tryptophan were detected by electron spin resonance (ESR) spectrometry. Solutions containing the amino acids and hydrogen peroxide were illuminated with ultraviolet radiation directly in the spectrometer cavity, where the hydroxyl radicals resulted from hydrogen peroxide photolysis. The ESR spectrum of the tryptophan radical was found to persist for several seconds after illumination ceased.

Other radicals were detected indirectly by passing a nitrous oxide-saturated solution containing various amino acids and p-nitrobenzoate through a cobalt-60 gamma-radiation field and into the ESR cavity. The p-nitrobenzoate ion was reduced to its radical anion in the presence of irradiated methionine, asparagine, or serine indicating the formation of radicals from these amino acids reacting to attach by hydroxyl radicals.

From the dynamic observation of optical absorption, transient species formed during flash photolysis of phenylalanine in the presence of hydrogen peroxide were ascribed to radicals of the cyclohexadienyl type. Coincidence of the optical absorption spectrum of this species with that of pulse-irradiated benzene in aqueous solution and their comparable reaction rates with molecular oxygen provide

evidence for this conclusion. In order to extend this type of observation, a compact flash lamp was assembled based on the z-pinch effect in gaseous plasma. This lamp provides a tenfold increase in light intensity and much more rapid deactivation of excited states in the source.

Damage Propagation with Biochemical Radicals

D. R. Kalkwarf

While the reactivities of radicals produced from water radiolysis have been studied extensively, it is also important to determine the capabilities of biochemical radicals to extend radiation damage by sequential and repetitive chain reactions.

Radicals were formed by irradiation of water-soluble biochemicals in the crystalline state and ESR spectra of the samples were measured to estimate the concentration and chemical structure of the radicals in this relatively stable environment. The irradiated solids were next dissolved rapidly in aqueous solutions of various diagnostic reagents where reaction with the reagent competed with radical-radical recombination and disproportionation. Control experiments with unirradiated solids and with irradiated samples which were dissolved in water prior to addition of the reagent served to establish which effects resulted from free-radical attack.

Free radicals derived from sugars and aliphatic amino acids generally acted as oxidizing agents toward the potent hydroxyl-radical scavenger, eriochlorine, although notable exceptions, e.g., sucrose and fructose, were found. Radicals from most sugars reduced flavin mononucleotide to its free-radical form but no amino acids were found to do so. Amino acid and sugar radicals exerted only a small effect on the viscosities of gelatin and polyvinylpyrrolidone solutions but were extremely potent in initiating polymerization of acrylamide. Significant viscosity changes in 1.0M acrylamide solutions were observed upon addition of glycine radicals to a concentration of only 10^{-5} M. Study of the reaction kinetics for this process showed it to be an accelerating chain reaction which eventually led to solidification of the solution. Examples of this type indicate that radicals temporarily stabilized in rigid biological tissue could act as delayed sources for inducing radiobiological damage.

PRESENTATIONS

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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 Neutron Activation Analysis

K. G. Pailthorp and H. E. Palmer

This study is concerned with the determination of elements of human body composition by in vivo neutron activation analysis after exposure to low levels of neutrons and the study of this elemental composition as a function of disease and disease state. In these studies various techniques of whole and partial body counting and scanning are used and significant improvements in these techniques and methods are undertaken.

A method for determining the relative loss or gain of total body calcium in humans was developed in conjunction with the Nuclear Medicine Group at the University of Washington cyclotron. Studies were made on three different cadavers to determine the precision of the method. The cadavers were each irradiated several times simultaneously with the irradiation of calcium nitrate standards. The ratio of the count rate of the cadavers to the count rate of the standards was

reproducible with a maximum coefficient of variation of 1.6% for all the measurements made on each cadaver.

The neutron fluence in these studies was ten times higher than that to be used on patients in order to minimize the error induced by counting statistics and to observe solely the inherent precision of the method.

With the neutron irradiation levels used for patient studies (a 0.2 rad radiation dose) a change of total body calcium greater than 3% can be detected with 95% confidence between two measurements made over a period of time. This is sufficient precision and sensitivity to assist the physicians in their studies of osteoporosis, bone wasting diseases, and their treatment. The method was approved by the Radiation Safety and Radioisotopes Committee of the University of Washington Medical School. Initial whole body calcium measurements on 20 patients have been made to date. Additional measurements to determine changes in total body calcium which have occurred in these patients will begin soon.

Instrumentation for Iron Uptake Measurements

H. E. Palmer and K. G. Pailthorp

The precision ^{59}Fe Whole Body Counter described in a previous report⁽¹⁾ was installed at the University of Washington and is being used on a continuous basis to study iron absorption in patients.

A proportional counter containing 100 ft of fine bore polyethylene tubing was developed in which the ^{55}Fe and ^{59}Fe content can be determined directly in blood which is drawn into the tubing. The counting efficiency is about 35 and 80% for the ^{55}Fe and ^{59}Fe , respectively, and their content can be determined simultaneously when both are present

in the blood. Development work is continuing on this counter; however, its potential value has already been demonstrated since at the present time all routine ^{55}Fe analyses in blood include steps of ashing, chemical separation and electrodeposition before counting can be undertaken.

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Neg 0694564-2

Enclosure for In Vivo Neutron Irradiation for Total Body
Calcium Analysis at the University of Washington

The Reproducibility of Measuring Calcium in a Cadaver

Cadaver	Ratio of Cadaver Count/ Standard Count	Ratio Average	Coefficient of Variation
No. 1	0.2052	0.2057	0.63%
198 lb	0.2071		
72 in.	0.2062		
	0.2063		
	0.2037		
No. 2	0.1864		
98 lb	0.1812	0.1841	1.4%
61 in	0.1846		
No. 3	0.1515		
154 lb	0.1501	0.1521	1.6%
	0.1547		

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RADIATION DOSIMETRY

L. A. Braby	W. C. Roesch
D. M. Fleming	L. H. Toburen
W. A. Glass	M. L. West
L. L. Nichols	W. E. Wilson

This study is primarily concerned with determining how to measure radiation to obtain correlation with biological effects. This is done by conceiving physical quantities that may serve the purpose, studying the principles that relate them to others previously known, determining how they are to be measured, and establishing how useful they are in correlating bioZogical phenomena.

In previous years the Radiation Dosimetry group and others had developed theoretical methods for calculating the probability density of the energy losses suffered by a charged particle while passing through a small volume. This group's principal activity this year has been the extension of these methods to calculating densities of quantities such as the energy deposited within the volume, the ionization produced there by the particle, or the ionization after multiplication in a proportional counter. The problem that had to be overcome was that, while the possible increments to the energy-loss are the same everywhere along the track of the particle, the possible increments to the quantities just mentioned differ from point to point along the track. Formulas have been developed that allowed for the point-to-point variation. Fortunately, the

group was also able to show that, if the correct average of the possible increments along the track of the particle were used, the same fast numerical methods developed for the energy-loss calculations can be used for these other quantities. Model calculations were carried out that explained the essential feature found earlier by Glass and Samsky for the energy deposition by protons in thin slabs. The theory was also used in analyzing the data obtained by Glass for protons passing at different distances from the center of a small volume.

Additional data for protons passing at different distances from the center of a small volume have been obtained for 4 MeV protons.

The limitations of the proportional counter techniques for microdosimetry measurements have been critically evaluated. The conclusion from this study is that proportional counters will probably be limited

to volumes larger than about 0.2 micrometer diameter sites (unit density material). This limitation is brought about because the finite volume of the avalanche region in the proportional counter becomes an appreciable part of the total volume of the absorbing site. The counter is therefore, no longer "proportional."

Measurements of the heat defect for 1.7 MeV protons on several materials were made using calorimetric techniques. Studies were completed for linear polyethylene, polystyrene, and polystyrene plus 20% carbon, and preliminary measurements were made on a sample of graphite provided by the National Bureau of Standards. Further measurements will be made on graphite and the results for all samples will be published elsewhere. These direct determinations of the effect of chemical reactions will reduce greatly the uncertainty in calorimetric measurements of dose rates.

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RADIATION PHYSICS

L. A. Braby	W. C. Roesch
D. M. Fleming	L. H. Toburen
W. A. Glass	M. L. West
L. L. Nichols	W. E. Wilson

This study is primarily concerned with how radiation produces effects in matter, particularly biological effects. This is done by studying the initial interactions by which ionizing radiations transfer energy to matter and the subsequent transport and degradation of that energy. The study includes determination of the partition and distribution of the energy in the radiation field and in the irradiated medium and determination of rates and mechanisms of energy transferred and transported.

Measurements are being made of the differential cross section for ejection of electrons by fast proton impact on gas targets. Cross sections, differential in electron emission angle and energy, were obtained for N_2 at nine angles between 20 and 130 degrees, and for five proton energies between 300 keV and 1.7 MeV. The cross sections, integrated over the emission angles, were compared to cross sections calculated from binary encounter theory. Good agreement is found except for electrons ejected with velocities approximately equal to the incident proton velocity. For electrons with these velocities, the calculated cross sections are about one half of the measured values. Similar results were obtained for electrons ejected from neon by 1.4 MeV protons and

also for 1.0 MeV protons on molecular hydrogen. These later results were compared to cross sections calculated by Inokuti using the Born approximation. The Born approximation gives cross sections also smaller than measured values in the region of the electron spectrum where the ejected electron and proton velocities are similar. Discrepancies of this type have been suggested by Oldham to be caused by interactions between the outgoing electrons and protons. This interaction is not properly accounted for in the Born approximation as generally applied.

Studies on the luminescence kinetics of organic liquids excited by ionizing radiation are in progress. Such studies provide useful information about elementary processes occurring in liquid scintillators during the formation and decay of

excited states. A positive ion accelerator has been converted to provide 100 keV electrons and a technique for measuring fluorescence lifetimes under y-ray excitation has been developed to complement our proton excitation measurements. With these three sources of excitation, we are able to study the time dependence of the scintillator response as a function of specific energy loss of the incident irradiation. Since all measurements can be made on the same sample at one time, the problems of unequal sample purity and environment can be eliminated. The single-photon technique used in the proton and y-ray excitation measurements has the advantage of allowing accurate measurements to be made on weakly emitting samples and, although this technique requires the use of carefully designed electronics, the ultimate time resolution is set by the statistical dispersion ($<0.5 \times 10^{-9}$ sec) of the phototube. Much effort has been directed this year toward achieving this limit. A new technique of constant fraction of pulse height has been used in triggering circuits to reduce the phototube time jitter to less than 0.5 nsec. The proton pulse width (chopped beam profile) has been reduced to less than 0.3 nsec and computer programs have been developed for unfolding the systems response function (proton pulse width plus phototube time jitter) from the observed emission curves.

A study has been made of the luminescence from benzene at room temperature upon excitation by electrons and protons. In addition to the singlet monomer fluorescence previously unobserved under electron and proton bombardment in liquid benzene, several fluorescing radiolysis products were observed. By using a flowing liquid target, most of the radiolysis-product emission was eliminated and thus spectra consistent with those reported for ultra violet excitation was obtained. Results suggest that the primary emission from benzene is the same for the various modes of excitation (i.e., via proton, electron, and photon irradiation) and since the singlet spectra are present under electron and proton bombardment, the proposed mechanisms of excimer formation via ion recombination is no longer necessary to explain the observed emission.

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8	<u>AEC Richland Operations Office</u> L. C. Brazley N. W. Fraser W. E. Lotz C. L. Robinson M. W. Tiernan D. G. Williams Technical Information Library (2)		
3	<u>Atlantic Richfield Hanford Company</u> D. J. Brown R. E. Tomlinson ARHCO Files		

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C. L. Simpson
A. J. Stevens
M. F. Sullivan
W. L. Templeton
R. C. Thompson
C. J. Touhill
C. M. Unruh
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E. E. Voiland

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