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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1971
TO THE USAEC DIVISION
OF BIOLOGY AND MEDICINE

VOLUME II: PHYSICAL SCIENCES
PART 2. RADIOLOGICAL SCIENCES

MAY 1972

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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1971
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

May 1972

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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-1650 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-1651 also in two parts: Atmospheric Sciences, Part 1; Radiological Sciences, Part 2.

FOREWORD

This report to the USAEC Division of Biology and Medicine covers work done in the fields of environmental and radiological chemistry, radiation instrumentation, radiation dosimetry, and radiation and health physics. It continues a series of reports on work initiated in 1959. Special effort has been made as last year 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:

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HW-73337	Period Ending December 31, 1961
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BNWL-1307 Part 2	Period Ending December 31, 1969
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TABLE OF CONTENTS

FOREWORD.	iii
<u>RADIOLOGICAL CHEMISTRY</u>	
<u>Inhalation Hazards to Uranium Miners.</u>	1
Elemental Content of Uranium Mine Atmosphere as a Function of Particle Size - J. A. Cooper, P. O. Jackson, T. M. Tanner and L. A. Rancitelli	1
Determination of Uranium Mine Aerosol Constituents Retained in Human Lungs - P. O. Jackson and J. A. Cooper	4
Characterization of Radon Daughter Components of Uranium Mine and Animal Test Chamber Aerosols - J. A. Cooper, P. O. Jackson, H. G. Rieck and R. W. Perkins	6
<u>Rates of Deposition of Airborne Radionuclides in the Alaskan Environment</u>	8
Radionuclides in the Alaskan Arctic Regions - C. E. Jenkins and James C. Langford.	8
Radionuclide Distribution in an Alaskan Forest Environment - C. E. Jenkins	11
Radionuclide Concentrations in North Pacific Salmon as a Function of Latitude - C. E. Jenkins and James C. Langford	12
Trace Element Content of Alaskan Caribou and Lichen - L. A. Rancitelli	15
<u>Physical and Radiological Chemistry of Ocean Solutions</u>	18
Latitudinal Variation of ⁷ Be Fallout in the Pacific Ocean - W. B. Silker	18
The Physical Form of Radionuclides in Pacific Ocean Water - W. B. Silker	20
The Relationship of ⁷ Be in Surface Water to that in the Water Column - W. B. Silker	21
Determination of Mercury in Seawater - D. E. Robertson	23
Multielement Studies of Micro and Macro Marine Organisms - L. A. Rancitelli	25
<u>Geochemical Oceans Sections Study (GEOSECS)</u>	
Northeast Pacific Ocean Station - D. E. Robertson	26
Northwest Atlantic Ocean Station - D. E. Robertson and W. B. Silker	26
<u>In-Cloud Scavenging</u>	29
Precipitation Scavenging of Tracers Released into Rainstorms Near Quillayute, Washington - J. A. Young, W. E. Davis, B. C. Scott, W. G. N. Slinn, T. M. Tanner, C. W. Thomas, J. M. Thorp and N. A. Wogman	29

Cosmogenic Radionuclide Production Rates in Argon from 3 to 19 km - J. A. Young, C. W. Thomas and N. A. Wogman	32
<u>Radioanalytical Procedure Development</u>	33
A ^{85}Kr Field Monitoring System and Its Application to Maximum Air Dose Measurements of a Radioactive Gas - J. D. Ludwick and P. W. Nickola	33
Analytical Procedure for Measuring Copper in Biological Material - C. W. Thomas and L. A. Rancitelli	34
Analytical Procedure for Measurement of ^{214}Pb and ^{214}Bi in Rainwater - C. W. Thomas, J. A. Young and N. A. Wogman.	35
Techniques Used to Stabilize Tracer Solution for Aerosol Generation - C. W. Thomas	36
<u>Radioactive Fallout Rates and Mechanisms</u>	38
Air Concentrations of ^{90}Sr and ^{55}Fe at Richland, Washington from 1963 to 1970 - C. W. Thomas and J. C. Langford	38
The Determination of Air-Sea Exchange and Oceanic Mixing Rates Using ^7Be During the BOMEX Experiment - J. A. Young and W. B. Silker	40
Trace Element Characterization of High Altitude Aerosol - T. M. Tanner and L. A. Rancitelli.	40
Comparison of Atmospheric Radionuclide Concentrations at Neah Bay and Richland, Washington - C. W. Thomas	42
Vertical Profiles of Radionuclide Concentrations in the Atmosphere - J. A. Young and N. A. Wogman	44
Atmospheric Fallout During 1971 - C. W. Thomas and J. A. Young	46
<u>Iron-55 and Lead in the Human Population of the World</u>	47
Iron-55 in Pacific Salmon, 1965-1970 - C. E. Jenkins and J. C. Langford	47
<u>Radiation Instrumentation - Radiological Chemistry</u>	50
The Performance and Application of a Versatile Ge(Li) - NaI(Tl) Coincidence-Anticoincidence Gamma-Ray Spectrometer for Environmental and Biological Problems - J. A. Cooper and R. W. Perkins	50
Natural Background Radiation in NaI(Tl) and Ge(Li) Gamma- Ray Spectrometers - N. A. Wogman and J. A. Cooper	52
Improved X-Ray Fluorescence Analysis of Environmental and and Biological Samples Using a High-Intensity Radioisotope Excitation - J. A. Cooper	55
X-Ray Fluorescence Determination of the Zinc to Copper Ratios in Fingernails for the Identification of Cystic Fibrosis - J. A. Cooper, C. E. Jenkins, J. C. Langford and N. A. Wogman	56
Transmission Properties of Specific Scintillator Reflective Coatings - R. L. Brodzinski	57
Computer System for Radionuclide Analysis - J. R. Kosorok	58

Multielement Analysis of Biological Reference Materials - L. A. Rancitelli	59
Characterization of Columbia River Basalt Flows from Major, Minor and Trace Element Considerations - L. A. Rancitelli	62
Trace Element Content of Aquatic Organisms from the Columbia River - L. A. Rancitelli and C. E. Cushing, Jr.	63
Radionuclide Distribution in Olympic National Park, Washington - C. E. Jenkins, N. A. Wogman and H. G. Rieck	64
Subcritical Multiplication of ^{252}Cf Neutrons and Its Application - N. A. Wogman and R. W. Perkins	66
<u>Tracer Studies in the METROMEX Experiment</u>	69
Preliminary Report on Experiments Conducted by Battelle- Northwest During METROMEX - J. A. Young, W. E. Davis, T. M. Tanner, C. W. Thomas and N. A. Wogman	69
Chemical Characterization of Urban Aerosol - L. A. Rancitelli and T. M. Tanner	72
<u>Radionuclide Decline in Columbia River Water.</u>	74
Transport and Depletion of Radionuclides in the Columbia River - D. E. Robertson, W. B. Silker and J. C. Langford	74
<u>RADIOLOGICAL PHYSICS</u>	
<u>Dosimetry of Radionuclides in Man</u>	81
Promethium - D. B. Shipler, J. E. Ballou, D. McConnon and I. C. Nelson	81
Plutonium - P. J. Dionne and I. C. Nelson	82
<u>Characterization of Radiation - Induced Free Radical Reactions in Aqueous Systems</u>	83
Chemical Reactivities of Radicals in Aqueous Solution - D. R. Kalkwarf, H. W. Fenrick and R. N. Diebel	83
Radical Reactions of Biochemical Membranes - D. R. Kalkwarf	84
Flash Photolysis of Nucleic Acid Components - W. D. Felix	85
<u>Nuclear Techniques in Medical Science</u>	87
Absolute Measurement of Total Body Calcium by In Vivo Neutron Activation Analysis - H. E. Palmer	87
In Vivo Production of ^{37}Ar and ^{41}Ar from Fast Neutron Irradiation of Calcium - H. E. Palmer	88
The Determination of Cadmium In Vivo by Measurement of Prompt Gamma Rays Following Neutron Capture - H. E. Palmer, L. W. Brackenbush and L. G. Faust	89
Radiation Instrumentation - K. L. Swinth	91
<u>Mechanisms of Environmental Exposure</u>	95

A Study of Seafood Consumption and ^{65}Zn Body Burden - T. H. Essig, G. W. R. Endres and J. F. Honstead	95
<u>Radiological Evaluation of Post Mortem Tissue Samples -</u> I. C. Nelson, K. R. Heid, P. A. Fuqua and T. D. Mahoney	98
<u>Radiation Dosimetry</u>	99
Energy Transport by Secondary Electrons - W. A. Glass	99
Hit Theory for Low-LET Radiation - W. C. Roesch	101
Theory of the Oxygen Effect - W. C. Roesch	103
Stopping Power Partition and Mean Energy Loss for Energetic Protons in Hydrogen - W. E. Wilson	105
<u>Radiation Physics</u>	108
Electron Emission Cross Sections - L. H. Toburen	108
Inner Shell Ionization and Fluorescent Yields - L. H. Toburen.	109
Fluorescent Decay of Organic Liquids - M. L. West.	110
Electron-Ejection Cross Sections for Several Representative Hydrocarbon Molecules - W. E. Wilson	111
<u>RADIOLOGICAL SCIENCES DEPARTMENT STAFF</u>	113
<u>Publications and Presentations</u>	115

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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 accurate characterization of the radionuclide, elemental and organic content of the uranium mine atmosphere and their correlations to lung exposure.

Elemental Content of Uranium Mine
Atmosphere as a Function of Particle
Size

J. A. Cooper, P. O. Jackson,
T. M. Tanner and L. A. Rancitelli

Atmospheric contaminants encountered in underground uranium mines are known to present serious occupational health hazards. Evaluation of the detrimental effects of the aerosol components requires that their elemental composition be known as a function of particle size.

Airborne particulates in several locations of an operating uranium mine were sampled with both Lundgren and Andersen impactors. The impactor stages on which particles of a specific size range collected were analyzed for their elemental content by both neutron activation analysis and isotopic X-ray fluorescence. Seventeen elements were measured and include Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Mn, Fe, Zn, Se, Sr, Pb and U. Additional analysis using long-lived neutron activation products are expected to yield results for another 15 elements.

The amount of material collected ranged from a few hundredths of a microgram of Mn to several hundred micrograms of Al. The weights of most of the elements were highest on the second stage of the Lundgren impactor (3 micron median diameter particles) and lowest on the fourth stage (0.3 micron median diameter particles). The Pb concentration, however, peaked on the first Lundgren stage (greater than 10 micron diameter particles) while the Zn concentration peaked on the fourth stage along with the carbon from diesel exhaust. This agreed well with the visual observation that most of the ore dust was deposited on the first two Lundgren stages while the diesel exhaust was deposited mostly on the fourth stage and backup filter. This suggests that the Zn may be associated with the diesel exhaust.

Inter-element ratios and correlations are being examined to determine whether specific elements are associated with a particle of a particular

size and to relate the mine conditions to those in the animal test chamber. Initial comparisons of elemental ratios showed some of them to vary considerably from one impactor stage to the next. Many of the elements varied by as much as two- or threefold relative to Al with the Zn to Al ratio increasing 25 times over the four stages of the Lundgren impactor. On the other hand, the Mg to Al ratio remained about constant over all four stages of the impactor and the backup filter. The Cl to Al ratio was also constant over the first three stages but increased by two- and tenfold on the fourth stage and backup filter, respectively. The Na to Al ratio followed a similar pattern but showed a smaller increase on the backup filter. The additional neutron activation analysis data and a complete correlation study of all of the elements will provide a greatly improved understanding of the contributions of various sources of uranium mine aerosols.

Neg 715759-1

Photograph of the Four Lundgren Impactor Stages and Backup Filter After Sampling About 1.8×10^4 Liters of Ambient Air and About 2.4×10^3 Liters of Exhaled Breath from Air in an Operating Uranium Mine

Determination of Uranium Mine Aerosol
Constituents Retained in Human Lungs

P. O. Jackson and J. A. Cooper

The inhalation of dust particles by mine workers is a contributing factor to pulmonary disorders. The potential hazards associated with dust inhalation is greater in uranium mines because of radon daughters which attach themselves to dust and the radiation from uranium and its long-lived daughters. The pulmonary deposition, location and retention of aerosol particles is a function of their size, density and associated aerodynamic properties.

The objectives of this study were to determine the fraction of the various types and sizes of aerosol particles which are retained in the respiratory system.

An instrument for collecting and sorting exhaled particulates has been developed and used in an operating uranium mine to determine pulmonary tract retention of stable and radioelement constituents of a uranium mine aerosol. It consists of an "air purification train" into which the subject exhales, and this air is rapidly carried through a Lundgren cascade impactor which separates particulates into 10, 3, 1 and 0.3 micron diameter groups. The impactor is followed by a backup filter which removes smaller particles. Sampling periods of two man-hours were employed for collecting sufficient material for elemental

analyses while five minute sampling periods in a rather poorly ventilated mine area were employed for short-lived radon daughter studies. The stages of the cascade impactor and the backup filter are analyzed for elemental content by neutron activation analysis and isotopic X-ray fluorescence. In the case of sampled mine air, the exhalates consisted of light-colored ore dusts observed on the first stages (larger sizes) and black carbon from diesel exhaust deposited mostly on the fourth stage and backup filter. Elemental analyses are not yet complete; however, the aluminum and vanadium, which were major components of uranium mine dust, and chlorine which was primarily associated with carbon loaded (diesel) stages in mine air samples are complete. The respiratory tract retention is higher for these elements when associated with particles 0.3 micron or greater than for those less than 0.3 micron. A higher fraction of aluminum was retained on the 1-3 micron particulates than on either 0.3 or 10 micron sizes. Chlorine was significant only on particulates less than 1 micron. Since it was higher in exhaled air than in normal mine air, it was apparently associated with biological components of breath. Analyses in progress will provide data for about 20 additional elements and permit a fuller evaluation of the respiratory tract fixation of dust constituents.

In the radon daughter experiments, the impactor stages and backup filter were counted with an alpha spectrometer a few minutes after sampling both exhaled breath and mine air. In both mine air and expired air the fraction of the radon daughter activ-

ity on particles larger than 0.3 micron was less than 0.3%. For both samples the radioactive equilibrium was essentially complete for particles collected on the backup filter, which was to be expected in the poorly ventilated study area.

Neg 715679-1

Apparatus for Measuring Atmospheric Components
Not Retained in Human Respiratory System as a
Function of Particle Size

Neg 715596-5

X-Ray Spectrum of the Elements in 3 to 10 Micron
Diameter Particles in the Atmosphere of an Operating
Uranium Mine Collected on the Second Stage of a
Lundgren Impactor

Characterization of Radon Daughter
Components of Uranium Mine and
Animal Test Chamber Aerosols

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

The major radiation hazard associ-
ated with uranium mining results from
pulmonary deposition of radon daugh-

ters. The radon daughter concentra-
tion must be known as a function of
particle size and time to relate the
conditions in our animal test cham-
bers to those in a uranium mine and
to evaluate the relative dose to the
lung. A sensitive research instru-
ment based on simultaneous alpha and
gamma-ray spectrometry was developed

and applied to the continuous measurement of the radon daughter concentrations as a function of particle size in the atmosphere of animal test chambers and in different locations of an operating uranium mine. Different Andersen impactor stages were used as pre-separators to select a given size particle which was then collected on a membrane filter and the activity measured. The pre-separation of large particles allowed continuous operation with a minimum of self-absorption effects by limiting the material collected on the filter to the fine particles with which essentially all of the radon daughter activity was associated. The amount of alpha self absorption was determined by comparing the intensity of the 7.69 MeV alpha peak of ^{214}Po with that of the 0.609 MeV gamma-ray peak from ^{214}Bi . The self absorption correction was less than

10% for collection periods of up to two hours in a dusty mine atmosphere.

The RaA (^{218}Po) and RaC¹ (^{214}Po) were measured directly from their 6.00 MeV and 7.69 MeV alpha peaks while the RaB (^{214}Pb) and RaC (^{214}Bi) were measured from their gamma-ray peaks at 0.295 and 0.352 MeV and 0.609 MeV, respectively. Equations were derived to relate a series of these measurements to the radionuclide concentration in the mine air as a function of time. The radon daughters associated with different sized particles were studied by using different Andersen impactor stages as pre-separators. The elemental content of the separated particles is being measured to establish the correlation with the radionuclide measurements and the distribution of elements on the stages of a Lundgren impactor.

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.

Radionuclides in the Alaskan
Arctic Regions

C. E. Jenkins and James C. Langford

Relatively few studies have been made of the behavior of radionuclides other than ^{55}Fe and ^{137}Cs in the Arctic regions. This program includes a study of the movement, uptake and relative behavior of 18 radionuclides from both natural and man-made sources. In addition to investigating the well known lichen-caribou-man (wolf) food web other possible avenues for uptake of radioelements by man are being studied to determine their relative importance. These measurements provide a basis for estimating the radiation dose received by resident Eskimos. These studies also provide basic information which will be extremely valuable in understanding deposition rates, movement, biospheric uptake and ultimate fate of toxic trace elements since many of the radioelements which are being investigated,

e.g., ^{65}Zn , $^{110\text{m}}\text{Ag}$, ^{125}Sb and ^{210}Pb , have stable toxic elemental counterparts from a wide spectrum of anthropogenic sources.

One important route by which radioelements may enter man is through his water supply. This can occur to a limited degree from drinking water but the major route is through aquatic organism concentration processes and the subsequent use of aquatic organisms as food. Measurements of the summer water supply of the Anaktuvuk Pass Eskimos, which consisted of a nearby creek (headwaters of the John River) showed radionuclide concentrations which were somewhat higher than those in the Columbia River upstream from the Hanford Plant. This pathway may be of greater importance in the Arctic region where lake and stream water are used directly for drinking purposes and local aquatic organisms are consumed.

Animals are the important source of radionuclides in the Alaska natives' diet. This is particularly evident from ^{55}Fe concentrations which have been measured. The yearly changes in ^{55}Fe concentrations since the 1961-1962 nuclear test series in various Arctic plants and ani-

mals are shown in the accompanying figures. Slow growing lichens show concentrations relatively constant since 1966 which may be partly due to additions from the Chinese nuclear testing while fast growing seasonal willows and grasses show sharp decreases through 1966.

Anaktuvuk Pass Water Sample^(a) - August 1, 1970
dpm 10^3

Radionuclide	Filters Suspended Sediments	Cation ^(b) Bed	Anion ^(c) Bed	Al Oxide Bed
^{22}Na	---	1.9 $\pm$ 0.35	---	---
^{40}K	3160 $\pm$ 200	204 $\pm$ 16	---	88.8 $\pm$ 16
^{54}Mn	14.9 $\pm$ 3	---	---	---
^{60}Co	8.73 $\pm$ 1	4.74 $\pm$ 0.45	---	1.30 $\pm$ 0.3
^{65}Zn	45.2 $\pm$ 6	67.2 $\pm$ 5	---	15.8 $\pm$ 3
$^{95}\text{Zr-Nb}$	108 $\pm$ 14	18.7 $\pm$ 1.8	29.3 $\pm$ 1.8	81.8 $\pm$ 14
^{106}Ru	---	43.1 $\pm$ 7	147 $\pm$ 6.4	446 $\pm$ 12
^{125}Sb	99 $\pm$ 5	34.9 $\pm$ 5	194 $\pm$ 4.7	577 $\pm$ 6
^{137}Cs	22.6 $\pm$ 2	16.0 $\pm$ 2.1	---	7.67 $\pm$ 2
^{208}Tl	7.83 $\pm$ 2	---	---	---
^{214}Bi	72.6 $\pm$ 6	53.9 $\pm$ 6	---	---

-
- a. 651 liters of creek water
 b. Two 1 in. thick cation beds
 c. One 1 in. thick anion bed
-

Neg 711947-8

^{55}Fe Specific Activity in Caribou and Reindeer

Neg 711947-6

^{55}Fe Specific Activity in Arctic Plants

Radionuclide Distribution in
an Alaskan Forest Environment

C. E. Jenkins

Information on radioelemental behavior in a forest system can add considerably to our knowledge of transfer and exchange processes. Preliminary investigations of radionuclide concentrations and the environmental factors involved in their large-scale movement were made in the Chugach Forest near Anchorage, Alaska. The upper Kenai River drainage basin has an annual precipitation rate of about 50 inches. This is about five times that at Anaktuvuk Pass. Since the deposition rates and subsequent movement of radionuclides is known to be related to the hydrology of an area, major differences in concentration and biogeospheric uptake between the tundra and forest environs were expected. The distribution and concentration of radioelements suggest that retention times in the flora component of the Alaskan forest are short in comparison to those of the Arctic tundra. Kenai lichen concentrations contain less long-lived and considerably more short-lived man-made fallout radionuclides than equivalent Anaktuvuk Pass flora.

Investigations of the Kenai soil component in the muskeg show a distinctive behavioral pattern of various radionuclides. Concentrations of the natural radionuclides ^{40}K , ^{232}Th , and ^{226}Ra are lower in the upper one inch of the heavy root zone than in the deeper zone. Other radionuclides have higher concentrations in the surface inch of soil. Thus, approximately 96% of the ^{60}Co , 67% of the ^{95}Zr - ^{95}Nb , 100% of the ^{106}Ru , 54% of the ^{125}Sb , 96% of the ^{137}Cs , and 100% of the ^{144}Ce are found in the first inch of soil. For ^{54}Mn the concentrations increase with soil depth for at least the first four inches. Zinc-65 concentrations are high at the surface, then drop but gradually increase again with depth.

River bottom sediments have predominantly those radioelements found in the upper inch of soil. The clay sediments have higher concentrations of ^{60}Co , ^{125}Sb and ^{137}Cs than the sand sediments. Very little ^{54}Mn was found in the river bottom sediments; however, it was shown to be present in the cationic fraction of water samples. These observations indicate that ^{54}Mn is rather highly mobile and reaches the plant root zone rapidly. It would thus be expected to find its way into plants through the roots.

* BOG-LIKE BOTTOM LAND

Neg 715720-1

Kenai Peninsula Soil Samples
Quartz Creek - August 1970

Radionuclide Concentrations in North
Pacific Salmon as a Function of
Latitude

C. E. Jenkins and James C. Langford

A study was initiated in 1967 to define the distribution of gamma-emitting radionuclides and ^{55}Fe in five mature salmon species sampled

at several locations along the western coast of the United States.

Immature salmon have also been measured to provide a basis for comparing river, estuarine, and oceanic food webs. In addition, information regarding the uptake of these various radioelements relative to their stable elemental counterparts is being

obtained. Radionuclide concentration in salmon appears to be the result of a complex interplay of several parameters. Among the most important are the oceanic migratory pattern of the individual salmon species, their life span, food web, and the relative dependence of their food web on elements supplied by river waters. Past and present studies confirm that some fallout radioelements are preferentially taken up by surface feeding organisms relative to their stable elemental counterpart by factors approaching 10^3 .

The group of fallout radioelements most effectively concentrated by salmon on a long-term basis are the elements ^{54}Mn , ^{55}Fe , ^{60}Co , and ^{65}Zn . These radioelements may be concentrated by factors of 10^3 to 10^5 over seawater concentrations. The highest ^{55}Fe and ^{54}Mn concentrations observed in salmon are in the northern latitudes. Fe-55 concentrations are 10 to 30 times higher in northern compared to southern salmon. This physical or chemical phenomenon appears related to the food web and for its relative dependence on fallout. Plots of ^{54}Mn , ^{60}Co , and especially ^{65}Zn concentrations in salmon versus latitude show the increase in concentration approaching the mouth of the Columbia. In 1968, Columbia River waters were approximately one, three, and several orders of magnitude higher in ^{54}Mn , ^{60}Co , and ^{65}Zn , respectively, than oceanic waters.

Since that time all of the Hanford reactors which discharge their cooling water directly to the Columbia River have been shut down and the concentration of the Columbia River radioelements has steadily declined.

A very slight dip in ^{55}Fe concentration and its specific activity which occurs in salmon caught near the Columbia River may be associated with the low ^{55}Fe specific activity of its waters. This is verified by the low ^{55}Fe specific activities in immature salmon which spent their lives in the river. The high concentrations of ^{60}Co and ^{65}Zn indicate the extent of the influence of the Columbia River waters. Ra-226 concentrations in immature Columbia River salmon show a dip similar to ^{55}Fe concentrations and specific activities. We have observed that ^{226}Ra which originates from soil or sediments is enriched in areas of high river output, in areas of ocean upwelling, and in areas where the seas are shallow. The Columbia River and the spring and summer upwelling along the Oregon coast may be responsible for high ^{226}Ra levels in salmon from the southern regions. High ^{226}Ra levels in Bering Sea salmon may be a result of shallow seas which enable ^{226}Ra from the sediments to reach the euphotic levels of the ocean more quickly. The concentrations of ^{137}Cs in salmon peak at the mid-latitudes where the fallout rate is the greatest and drop rapidly

southward, closely following concentrations of ^{137}Cs in the seawater.

Annual measurements of radionuclides in salmon from Kotzebue and the Columbia River have shown rapid declines. Kotzebue salmon show ^{55}Fe and ^{54}Mn declines that approximate the stratospheric input rate. Cs-137 levels are still rising, apparently due to its long half-life and resident time in the oceans. Co-60 levels

appear to be leveling off as would be expected for this intermediate half-life radionuclide. Columbia River salmon are showing rapid yearly declines in ^{54}Mn , ^{55}Fe , ^{60}Co and ^{65}Zn . The ^{55}Fe decline is similar to that observed for fallout. Mn-54 , ^{60}Co and ^{65}Zn are falling at a much more rapid rate, indicative of the Hanford facility shutdowns.

$^{55}\text{Fe}/\text{Fe}$ SPECIFIC ACTIVITY IN PACIFIC SALMON-1968

DISTRIBUTION OF ^{55}Fe IN PACIFIC SALMON-1968

DISTRIBUTION OF ^{54}Mn IN PACIFIC SALMON-1968

DISTRIBUTION OF ^{60}Co IN PACIFIC SALMON-1968

DISTRIBUTION OF ^{137}Cs IN NORTH AMERICAN SALMON IN 1968

DISTRIBUTION OF ^{226}Ra IN PACIFIC SALMON-1968

Neg 711283-3

Trace Element Content of Alaskan Caribou and Lichen L. A. Rancitelli

The transport and distribution of radionuclides in the Alaskan environment has received a great deal of attention due to the unusually high radionuclide concentrations in cer-

tain principal food chains. While significant advances have been made in understanding the transport of fallout radionuclides from the air through the marine and terrestrial food chains to the native population, the effects of dilution of these radionuclides by their stable element counterparts have received very

little attention. Thus, to help elucidate the mechanisms responsible for radionuclide transport through the Alaskan food chain, a study is being conducted of 15 elements: Ag, As, Br, Co, Cr, Cs, Fe, Hg, K, Na, Rb, Sb, Sc, Se and Zn, in lichen and caribou, important links in a major chain leading to man. The measurements are made by instrumental neutron activation analysis using 200 mg aliquots of caribou liver, muscle and kidney tissue and lichen samples which are simultaneously analyzed for their radionuclide content. The specific activity data obtained from the trace element study and the radionuclide analysis permit the importance of these radionuclides in the food web to be characterized.

Preliminary results of the trace element analyses of caribou reveal some trace element patterns similar to those observed in marine and aquatic organisms. The majority of the silver is contained in the liver. However, the high silver content of liver was not accompanied by high Se concentrations as previously noted in salmon. The caribou kidney and

liver contained far higher cobalt concentrations than the muscle tissue. The muscle tissue contained the highest levels of zinc, in marked contrast to marine organisms where the highest concentrations are found in the liver and kidney. The mercury content of the caribou tissue samples was below 0.5 ppm, and in muscle tissue was below 0.02 ppm on a fresh weight basis. The interpretation of the elemental analyses of lichen samples is difficult, since it is obvious from the Sc, Sb and Fe concentrations that a significant amount of soil material was contained in the samples. However, several important observations are apparent in spite of the soil component; the Se content (0.03 to 0.08 ppm) is below the range of 0.2 to 0.8 ppm recommended by the Research Council's Agricultural Board as necessary for domestic livestock food. Selenium is suspected to be an essential element and the effects of Se deficiencies on livestock are well documented. The mercury and arsenic contents of lichen are also low, 0.05 ppm and <0.5, respectively..

Trace Element Content in Alaska Lichen and Caribou
ppm Dry Tissue

Tissue	Ag	As	Br	Co	Cr	Cs	Fe	Hg	K	Na	Rb	Sb	Sc	Se	Zn
Lichen															
Sample #1	0.044	<0.20	0.38	0.078	0.27	0.036	160	0.014	490	39	1.1	0.012	0.063	0.034	9.0
Sample #2	0.042	<0.51	1.1	0.32	0.84	0.12	530	0.051	2,400	180	3.6	0.046	0.19	0.081	30
Caribou 1-69															
Flesh	<0.001		8.8	0.015	<0.094	0.083	140	0.035	19,500	1,300	32	0.00038	0.00023	0.35	130
Liver	0.25		13	0.46	<0.14	0.036	820	0.56	15,500	1,600	47	0.0045	0.00089	0.81	81
Kidney	0.0017		44	0.43	<0.24	0.095	150	2.2	16,200	5,800	44	0.0067	<0.00005	3.4	93
Caribou 2-69															
Flesh	<0.001		17	0.025	<0.12	0.11	180	0.093	14,800	1,500	42	0.00038	0.00029	0.48	220
Liver	0.16		18	0.22	<2.2	0.027	230	0.12	9,900	1,100	56	0.0030	0.00040	0.68	82
Kidney	<0.001		55	0.36	<0.17	0.083	260	0.45	14,800	4,300	63	0.0013	<0.00005	3.0	89
Caribou 3-69															
Flesh	<0.001		6.6	0.029	<0.084	0.11	140	0.020	17,600	1,000	40	0.0035	0.00032	0.47	220
Liver	0.28		24	0.41	<0.13	0.031	520	0.51	14,500	3,200	34	0.0080	0.00044	0.80	82
Kidney	0.0047		30	3.0	0.93	0.084	240	1.8	16,100	3,900	48	0.11	0.019	3.7	100
Caribou 4-69															
Flesh	<0.001		9.2	0.022	<0.19	0.22	170	0.061	13,800	1,000	43	0.0022	0.00026	0.34	120
Liver	0.18		16	0.20	<0.14	0.078	290	0.11	12,000	1,600	56	0.0042	0.00016	0.56	53
Kidney	0.0022		43	0.69	<0.25	0.13	220	0.24	13,900	3,400	75	0.0099	<0.00005	3.7	89
Caribou 6-69															
Flesh	<0.001		7.7	0.016	<0.12	0.043	160	0.076	15,800	1,100	14	0.0034	0.00016	0.37	93
Liver	0.30		15	0.31	<0.14	0.019	310	0.23	12,400	1,500	20	0.0051	0.00061	0.86	85
Kidney	<0.001		46	0.35	<0.16	0.064	140	1.3	14,900	5,500	25	0.00046	0.00005	3.9	110

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 and 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.

Latitudinal Variation of ^7Be Fallout in the Pacific Ocean

W. B. Silker

Data presented in another section of this report show a relationship between the total ocean water column inventory of ^7Be and its surface concentration. A similar relationship was found for $^{95}\text{Zr-Nb}$. Thus the surface water concentrations of these radionuclides can be used to evaluate both their transfer rate across the air-sea interface and the total input from recent fallout.

The accompanying figure shows the surface water concentrations of ^7Be in samples collected on three north-south transects of the Pacific. The samples collected two years apart along the eastern Pacific Ocean boundary show remarkable consistency in their latitudinal ^7Be radionuclide distribution. Of particular interest are the very high concentrations found at about 10°N latitude along these lines and the minor maximum at the same latitude in the mid-ocean track. The air-to-sea transfer rate near the

eastern Pacific Ocean boundary is some 50-fold greater at 10°N latitude than at the equator. This latitudinal distribution differs drastically from those found in the Health and Safety Laboratory 80th Meridian studies for ^{90}Sr or ^{137}Cs , which both increase regularly from minimum equatorial values to a maximum at $40\text{--}50^\circ\text{N}$. The ^7Be maxima observed at 10°N are probably at least partially due to wet deposition by precipitation. Total annual rainfall, both along the 160th meridian and at Pacific Coast stations, exhibits a very pronounced maximum between $5\text{--}10^\circ\text{N}$, decreasing to a minimum at both the equator and mid-latitudes. Weather satellite data reveal persistent and highly developed cloud cells over the eastern Pacific with maximum cloudiness about 8°N and decreasing cloudiness both north and south. The southern hemispheric data also are interesting because of the much higher concentrations shown for mid-ocean samples when compared with those taken along the eastern margin. Surface samples taken on a track between $25\text{--}30^\circ\text{S}$ showed quite high concentrations

west of 130°W longitude with decreasing values to the eastward. This also is the western boundary of the cold Peru Current. Almost desert conditions of low rainfall prevail in the eastern equatorial south Pacific because of the lack of water transfer from the cold ocean surface to the atmosphere.

The excellent agreement between surface water radionuclides and the rainfall and cloud distributions

strongly suggests wet deposition to be an important mode of transfer of radionuclides from the atmosphere in the regions of high ^7Be concentration. The important fact from these data is that certain areas of the tropical ocean receive much higher quantities of fallout, at least at certain times of the year, than had previously been recognized.

Neg 715485-1

^7Be Concentrations in Pacific Ocean Surface Water

The Physical Form of Radionuclides
in Pacific Ocean Water

W. B. Silker

The application of ^7Be and fission products as tracers of oceanographic mixing processes requires that they act conservatively, being distributed within the water column only by the physical forces acting on the water mass. Many investigators have either theorized or found by measurement that many fission products, including $^{95}\text{Zr-Nb}$, ^{106}Ru and ^{144}Ce , were largely associated with oceanic particulates, while others, conversely, found large percentages to be "soluble."

In a study of this question, data from 120 samples of North Pacific Ocean water taken from surface to 100 meter depths were analyzed as a function of depth with respect to the radionuclide fraction associated with the particulates. These samples were taken over a large oceanic area extending from the equator to the Gulf of Alaska and

the continental coastline to approximately 180°W during 1968-1971. In the following table the percentages of ^7Be , $^{95}\text{Zr-Nb}$, ^{106}Ru and ^{144}Ce which are retained on 0.3μ pore size filters are presented as a function of depth. The percentages associated with particulates are given in terms of the median, the average and the maximum.

These data show that the fraction of these radionuclides associated with particulate material is generally 10% or less. They also show that a rather constant fraction of these radionuclides is associated with the particulate phase throughout the top 100 meters of the water column. Therefore, the particulate fraction, regardless of physical form, is apparently behaving within this 100 meter deep water column in the same manner as a "soluble" radionuclide. The radionuclides of this group therefore, are excellent tracers for measurement of vertical diffusion and advective transport in the open oceans.

Percentage of Radionuclides with Particulates in Seawater						
Median, Average, Maximum						
Depth (meters)	0	15	30	50	75	100
No. of Samples	32	19	26	16	15	12
<u>Isotope</u>						
^7Be	5,9,27	11,11,24	10,8,22	7, 9,19	7, 8,19	0,10,39
$^{95}\text{Zr-Nb}$	7,7,16	8,10,17	8,7,18	7, 8,17	9,11,25	11,11,25
^{106}Ru	2,3,12	1, 2, 5	2,2, 9	1, 1, 3	1, 2,15	0, 0, 2
^{144}Ce	0,7,37	0, 7,26	5,7,28	10,13,30	5, 7,29	7, 8,32

The Relationship of ^7Be in Surface
Water to That in the Water Column

W. B. Silker

Beryllium-7, produced in the atmosphere by cosmic-ray spallation of nitrogen and oxygen, may serve as a tracer for measurement of vertical eddy diffusion rates within the upper mixing layer of the oceans. Owing to its relatively short half life ($T_{1/2} = 53$ days), it is possible to obtain the inventory of ^7Be by sampling to limited depth, and this inventory represents the steady state addition by transfer from the atmosphere to the sea surface. Thus, by determination of the concentrations contained by a number of water columns, representative of the total ocean, the cumulative input of atmospheric aerosols to the ocean may be estimated. Limitations of time and logistics have precluded such an ambitious project. If, however, a relationship exists between the concentration of ^7Be in surface water and the water column inventory, the estimation of the transfer across the air-sea interface will be greatly simplified.

The ^7Be production rate varies with both latitude and altitude. Surface air concentrations vary seasonally about a factor of three with the northern hemisphere maximum occurring during June and the minimum in December. If it is assumed that the transfer of ^7Be across the air-sea interface is proportional to the atmospheric concen-

tration, then its concentration in surface water should closely parallel the seasonal fluctuations in the atmosphere, and the water column inventory would be a function of the total transfer during the preceding several months.

Data derived from twenty-one profiles sampled from 1968-71 north of the equator in both the Atlantic and Pacific oceans are shown in an accompanying figure. These data are presented as the variation with time of year of the ratio of the surface concentration of ^7Be to its water column inventory. A strong seasonal change of this ratio is apparent. Highest ratios occur during July and August following the period of highest air concentrations, and hence the highest input, when the total water column inventory of ^7Be is at its highest value. Low ratios occurring during the early spring months reflect the combination of both minimum airborne radionuclide concentration and the high ^7Be inventory accumulated during preceding higher input periods.

The deviation of the individual values from the curve in the figure was 11.2% at the 95% confidence level. From these data it appears that the input rate of ^7Be , and presumably of other atmospheric contaminants, can be determined within about 11% simply by monitoring the surface water concentration. This is remarkable, considering that the surface concentrations ranged from 200-900 dpm/m³, and that

the profiles were sampled over a three-year period across a wide range of longitude and from 8°-45° north latitude.

Future work is required to determine the shape of the curve during months that have not yet been sampled, and

perhaps more detailed depth sampling is needed to define any fine structure in the depth distribution of ^7Be which would permit a more accurate assay of the water column inventory.

Neg 720069-1

Seasonal Change of the Ratio ^7Be Water Column
Inventory to Its Concentration in Surface Water

Determination of Mercury in Seawater

D. E. Robertson

Because of the great concern for mercury contamination of the marine environment by man's activities, it is essential that the mercury concentrations and distribution in the ocean be determined. Since the mercury levels in the oceans appear to range between 0.01 to 0.3 $\mu\text{g Hg/l}$, it is necessary to preconcentrate the mercury to meet instrument sensitivity and to minimize interferences from other elements. A rapid, simple procedure for quantitatively measuring mercury in 100-200 ml of seawater has been developed, based on its amalgamation with copper metal and subsequent analysis by flameless atomic absorption spectrophotometry. Seawater (usually acidified to pH 1.5 with high purity hydrochloric acid) is passed through a 1.0 cm diameter quartz tube containing a 1.5 cm long column of 30 mesh, granular, copper metal* at a flow rate of 3 ml per minute. The copper is rinsed with a few ml of purified water and a few ml of acetone, then thoroughly dried by passage of a stream of clean air. The quartz tube is placed in a furnace preheated to 250°C.

* *Minco copper metal, fine granular; Midwest Instrument Co., Delafield, Wisconsin.*

The volatilized mercury is swept, with a stream of N_2 gas, through an optical cell placed in the light beam of an atomic absorption spectrophotometer. Mercury is measured by absorbance at the 2536.5 Å resonance line. The yield and precision of this method, determined by six replicate analyses, with radioactive $^{203}\text{Hg}^{++}$ tracers, was $96.7 \pm 0.9\%$. Preliminary analysis of deep Pacific Ocean water collected several hundred miles off San Diego, California gave mercury values ranging between 0.17-0.20 $\mu\text{g Hg/l}$.

When methyl ^{203}Hg chloride tracers were used to evaluate the procedure, a yield of only $4.0 \pm 0.2\%$ was recorded. Thus, this analytical method is almost completely insensitive to methyl mercury. It has been shown, however, that methyl mercury can be readily photo-oxidized and subsequently retained on the granular copper metal column. Inorganic and methyl mercury can thus be separately measured by first extracting and measuring the inorganic mercury. The effluent seawater is then subjected to ultraviolet irradiation to oxidize the methyl mercury. Then the seawater sample is reanalyzed for mercury. Mercury observed in the second analysis is thus a measure of the Hg in the methyl mercury form and possibly in other organo-metallic compounds.

Storage of seawater samples presents numerous problems due to losses of certain elements by adsorption onto container surfaces. Storage conditions for seawater collected for mercury analysis were evaluated by spiking seawater with $^{203}\text{Hg}^{++}$ and methyl ^{203}Hg chloride tracers and storing at pH 1.6 and 8.0 in polyethylene bottles. At pH 8.0 both the $^{203}\text{Hg}^{++}$ and methyl ^{203}Hg chloride are rapidly and nearly quantitatively lost from solution by

adsorption onto the polyethylene containers. However, when the pH is reduced to 1.6 with hydrochloric acid no serious adsorption of these tracers occurs. Our standard method of handling seawater samples consists of acidifying the seawater to pH 1.5 to 1.6 at collection time with high purity hydrochloric acid and storing in acid-cleaned polyethylene bottles; thus the adsorption problem is minimized.

Neg 713989-7

Adsorption of $^{203}\text{Hg}^{++}$ and Methyl ^{203}Hg Chloride
From Sea Water at pH 1.6 and 8.0
onto Polyethylene Container Surfaces

Multielement Studies of Micro and
Macro Marine Organisms

L. A. Rancitelli

The trace element content of Pacific salmon, phytoplankton, zooplankton and macro algae has been extensively characterized in an attempt to understand the pathways and biological uptake of radionuclides and pollutants in the marine environment. Samples of muscle and liver from five species of Pacific salmon have been analyzed by a multi-element technique of instrumental 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 thirty-five salmon. Relationships between the alkali metals K, Rb and Cs were established which indicate that the radiologically important element Cs is preferentially assimilated relative to K and Rb. The Rb to K ratios in salmon tissue and seawater are identical. The simultaneous determination of 14 elements in each tissue specimen permitted a statistical analysis of the data which revealed other trace element correlations.

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 significance level of 10^{-6} for the number of samples considered, indicating only one chance in a million of a random correlation. 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. A comparison of the elemental abundances in phytoplankton, zooplankton, macro algae and salmon muscle indicates none of the elements considered in this work are substantially concentrated in the higher trophic levels of the marine food chain.

GEOCHEMICAL OCEAN SECTIONS STUDY (GEOSECS)

The multi-institutional GEOSECS program is designed to measure a large group of ocean constituents and hydrographic parameters during transects of the world's major oceans, with the aim of better understanding the geochemistry and mixing dynamics of the oceans. Two preliminary intercalibration and testing cruises, one in the N.E. Pacific and another in the N.W. Atlantic, have been completed in preparation for the main GEOSECS expeditions which will begin this summer in the N.W. Atlantic Ocean. Battelle-Northwest has participated in both of these intercalibration cruises, measuring trace element, ^7Be and fallout radionuclide concentrations in depth profiles. During the GEOSECS program these measurements will help characterize the ocean's geochemistry, determine the worldwide oceanic deposition of radionuclides and define the vertical mixing rates above, through and below the main thermocline. The results of these preliminary studies of trace element and radionuclide measurements are summarized.

Northeast Pacific Ocean Station

D. E. Robertson

The results of trace element measurements and interlaboratory comparisons at the Pacific pre-GEOSECS station have been reported in detail in: Spencer, D. W., D. E. Robertson, K. K. Turekian and T. R. Folsom. Trace Element Calibrations and Profiles at the Geosecs Test Station in the Northeast Pacific Ocean. J. Geophys. Res. 75, 7688-7696, Dec. 20, 1970.

Northwest Atlantic Ocean Station

D. E. Robertson and W. B. Silker

In August, 1970 a second pre-GEOSECS (Geochemical Ocean Sections Study) intercalibration and testing cruise was conducted in the Northwest Atlantic Ocean aboard the R/V Knorr. A station at 36°N latitude, 68°W longitude was occupied

during which vertical sampling was conducted to determine the depth distribution of various ocean constituents. The vertical distributions of ^7Be , $^{95}\text{Zr-Nb}$, ^{106}Ru and ^{144}Ce were measured utilizing large-volume sampling techniques and high sensitivity gamma-ray spectrometry. These measurements have been summarized in the 1970 Annual Report BNWL-1551, Vol. II, Part 2.

Sea water samples were also collected for interlaboratory comparisons of trace element concentrations at 29 depths between the surface and the ocean floor in modified 30 liter Niskin bottles. The unfiltered seawater samples were acidified to about pH 1.5 with high purity HCl and stored in polyethylene bottles. Aliquots of the seawater samples were divided among Battelle-Northwest Laboratory, Woods Hole Oceanographic Institution and Yale University.

Instrumental neutron activation analysis was employed to measure the concentrations and depth distributions of Zn, U, Co, Cs, Sb and Sc. This method consists of evaporating 25 ml aliquots of seawater and neutron activating the dried salts, together with appropriate standards, to an integral thermal neutron exposure of about 10^{18} n/cm². The samples were stored for a month and then counted on a multidimensional gamma-ray spectrometer and Ge(Li) diode detector. The samples were analyzed in duplicate about one month apart. The U, Sb and Cs were

very homogeneously distributed throughout the entire water column. Unlike the Pacific pre-GEOSECS station, the Zn, Co and Sc distributions showed no systematic variation with depth, even though large scattered variations were observed. It appeared that several of the Co values represented contamination introduced during the collection or storage of the samples. The results of the Woods Hole and Yale groups are not yet available, so no interlaboratory comparisons of the Atlantic Ocean samples can be reported at this time.

Trace Elements in Atlantic Ocean Waters
Pre-GEOSECS R. V. Knorr Cruise #9
Station 21 36°N Latitude 68°W Longitude
August 1970

DEPTH (METERS)	BNW SAMPLE NO.	WHOI		SALINITY	CONCENTRATIONS IN µg/LITER											
		SAMPLE	IDENT.		Sc	Co	Sb	Cs	U	Zn						
1	2	CAST 8#	519	36.075	0.0006 0.0007	0.0007	0.013 0.016	0.014	0.19 0.19	0.29 0.31	0.30	3.7 3.6	3.6	1.2 1.4	1.3	
90	3	CAST 8#	561	36.536	0.0004 0.0003	0.0004	0.147 0.143	0.145*	0.24 0.23	0.29 0.30	0.30	3.6 3.4	3.5	2.1 2.3	2.2	
190	18° SARGASSO SEA WATER	4	CAST 8#	564	36.550	0.0005 0.0006	0.0006	0.010 0.012	0.011	0.19 0.18	0.29 0.30	0.29	3.4 3.4	3.4	1.2 1.3	1.2
290		1	CAST 8#	511	36.524	0.0010 0.0010	0.0010	0.013 0.015	0.014	0.21 0.21	0.29 0.30	0.30	3.9 3.5	3.7	3.6 3.5	3.5
390		5	CAST 8#	565	36.500	0.0004 0.0004	0.0004	0.010 0.019	0.015	0.18 0.22	0.28 0.31	0.29	3.2 3.6	3.4	1.4 1.5	1.4
490		6	CAST 8#	566	36.469	0.0007 0.0009	0.0008	0.456 0.455	0.455*	0.22 0.24	0.28 0.28	0.28	3.3 3.3	3.3	2.6 2.5	2.5
758	10	CAST 15#	569	35.886	0.0007 0.0005	0.0006	0.011 0.009	0.010	0.20 0.19	0.29 0.29	0.29	3.3 3.1	3.2	1.2 1.5	1.4	
960	O ₂ MIN.	8	CAST 15#	567	35.210	0.0008 0.0007	0.0007	0.013 0.015	0.014	0.21 0.21	0.28 0.29	0.28	3.5 3.5	5.0 5.0	5.0	
1263		12	CAST 15#	572	34.991	0.0006 0.0006	0.0006	0.013 0.015	0.014	0.18 0.19	0.28 0.29	0.28	3.4 3.4	1.0 2.0	1.5	
1515		9	CAST 15#	568	34.983	0.0007 0.0007	0.0007	0.024 0.024	0.024	0.21 0.23	0.28 0.29	0.29	3.3 3.6	1.5 1.6	1.6	
2020	NORTH ATLANTIC DEEP WATER	11	CAST 15#	571	34.968	0.0004 0.0006	0.0005	0.015 0.014	0.014	0.20 0.19	0.29 0.30	0.29	3.4 3.3	1.4 2.1	1.8	
2525		7	CAST 15#	510	34.957	0.0008 0.0009	0.0008	0.051 0.051	0.051	0.19 0.20	0.25 0.26	0.26	3.2 3.1	2.9 3.0	3.0	
2983		13	CAST 20#	507	34.947	0.0009 0.0011	0.0010	0.019 0.023	0.021	0.24 0.27	0.26 0.30	0.28	3.1 3.5	2.8 3.3	3.0	
3232		14	CAST 20#	522	34.929	0.0010 0.0010	0.0010	0.016 0.017	0.016	0.26 0.24	0.27 0.27	0.27	3.2 3.2	4.5 4.4	4.4	
3667		16	CAST 20#	539	34.918	0.0009 0.0012	0.0010	0.035 0.042	0.039	0.18 0.27	0.25 0.30	0.28	3.1 3.5	2.0 3.0	2.5	
4160	ANTARCTIC BOTTOM WATER	15	CAST 20#	537	34.895	0.0010 0.0010	0.0010	0.011 0.015	0.013	0.24 0.21	0.28 0.27	0.27	3.4 3.3	2.4 3.1	2.7	
4509		17	CAST 20#	541	34.893	0.0010 0.0010	0.0010	0.022 0.027	0.024	0.19 0.28	0.25 0.30	0.28	3.2 3.7	1.4 2.7	2.1	
4780		18	CAST 20#	542	34.890	0.0010 0.0010	0.0010	0.008 0.011	0.010	0.17 0.26	0.26 0.30	0.28	3.1 3.7	2.5 2.9	2.7	
3998		22	CAST 35#	557	34.900	0.0010 0.0010	0.0010	0.008 0.010	0.009	0.18 0.19	0.28 0.26	0.27	3.5 3.2	1.1 1.3	1.2	
4748		19	CAST 35#	502	34.886	0.0010 0.0010	0.0010	0.008 0.012	0.010	0.19 0.20	0.29 0.28	0.29	3.6 3.2	1.9 1.5	1.7	
4823	ANTARCTIC BOTTOM WATER	20	CAST 35#	548	34.885	0.0010 0.0010	0.0010	0.008 0.009	0.009	0.18 0.20	0.28 0.29	0.29	3.5 3.5	3.1 3.0	3.1	
4870		21	CAST 35#	555	34.884	0.0008 0.0007	0.0008	0.007 0.007	0.007	0.22 0.21	0.31 0.28	0.29	3.6 3.4	1.2 1.5	1.4	
4923		24	CAST 35#	560	34.883	0.0009 0.0010	0.0009	0.031 0.043	0.037	0.18 0.20	0.27 0.26	0.27	3.5 3.6	2.7 2.5	2.6	
4963		23	CAST 35#	559	34.882	0.0010 0.0010	0.0010	0.008 0.009	0.008	0.19 0.17	0.28 0.27	0.28	3.5 3.2	1.5 1.6	1.5	
4998		25	CAST 35#	562	34.882	0.0004 0.0004	0.0004	0.005 0.008	0.006	0.25 0.23	0.29 0.28	0.29	3.4 3.3	1.7 1.6	1.7	
4919		29	CAST 41#	547	34.886	0.0010 0.0010	0.0010	0.018 0.016	0.017	0.21 0.23	0.29 0.29	0.29	3.5 3.4	1.8 2.5	2.1	
4944	ANTARCTIC BOTTOM WATER	28	CAST 41#	546	34.886	0.0010 0.0010	0.0010	0.018 0.010	0.014	0.22 0.23	0.29 0.27	0.28	3.6 3.4	1.4 1.8	1.6	
5044		26	CAST 41#	538	34.887	0.0010 0.0010	0.0010	0.018 0.014	0.016	0.25 0.24	0.29 0.28	0.29	3.5 3.4	1.6 2.2	1.9	
5069		27	CAST 41#	543	34.903	0.0009 0.0010	0.0010	0.017 0.018	0.017	0.23 0.23	0.28 0.28	0.28	3.2 3.3	3.6 4.4	4.0	

* SEA WATER SAMPLES OBVIOUSLY CONTAMINATED DURING COLLECTION AND STORAGE

Hoko. When a water droplet or ice crystal begins to grow primarily by coalescence, its radius to volume ratio decreases and the rate of capture of newly formed cosmogenic radionuclides by the growing hydrometeor should rapidly become negligible compared to the cosmogenic radionuclide activity of the hydrometeor. The cosmogenic radionuclides on the hydrometeor should then begin to decay away and the $^{39}\text{Cl}/^{38}\text{Cl}$ ratio should increase with time at a rate determined by the ^{38}Cl and ^{39}Cl half-lives. Therefore, the hydrometeor lifetime can be calculated from the $^{39}\text{Cl}/^{38}\text{Cl}$ ratio of the precipitation, assuming that the $^{39}\text{Cl}/^{38}\text{Cl}$ activity ratio on the droplet or ice crystal was at the equilibrium value of 1.5 at the beginning of growth by coalescence. The hydrometeor lifetimes calculated in this manner varied from nearly zero to 99 minutes, and did not appear to be significantly different between Quillayute and Hoko. The time showed a tendency to decrease throughout the storm.

On April 16, 1971 indium, gold, iridium, and silver iodide were released into precipitating clouds near the Quillayute airport. Indium and

silver iodide were released by means of pyrotechnic flares, while the iridium and gold were released by burning heptane solutions of iridium and gold in aerosol generators. Rain samples were collected in the same manner as in the previous tracer release of March 11, 1971, except that a slightly larger network of 90 samplers was used. The samples have been analyzed by neutron activation for indium and gold. The samples are being analyzed at the present time for iridium and silver.

Large volume rain samplers were also collected for cosmogenic radionuclide analysis at Quillayute and Camp Hoko on April 16, 1971. Both the ^{38}Cl and ^{39}Cl concentrations and the ^{38}Cl to ^{39}Cl ratios were appreciably higher at Hoko than at Quillayute. The hydrometeor lifetimes, calculated in the manner described above varied from nearly 0 to 58 minutes at Quillayute and from 41 to 94 minutes at Hoko. Of four rainstorms studied in early 1971, this was the only one in which there were significant differences in the cosmogenic radionuclide concentrations between Quillayute and Hoko.

Neg 720282-4

Rain Collection Network on the Olympic Peninsula
of Washington State

Cosmogenic Radionuclide Production

Rates in Argon from 3 to 19 km

J. A. Young, C. W. Thomas

and N. A. Wogman

For the past four years the concentrations of the cosmogenic radionuclides ^{24}Na (19 hr), ^{38}Cl (37 min) and ^{39}Cl (55 min) have been measured in air filter samples collected by aircraft. These radionuclides are produced at a relatively constant rate in the atmosphere 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 were lined with rubber meteorological balloons, filled with argon under pressure, and flown at altitudes ranging from 3 to 19 km. 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 NaI(Tl) multidimensional gamma-ray spectrometer. In addition to ^{24}Na , ^{38}Cl and ^{39}Cl , the cosmogenic radionuclides ^{18}F , ^{28}Mg , ^{34}Mg and ^{38}S were measured on some of the 15 to 19 km flights.

The measured ^{24}Na production rates were 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 and ^{39}Cl production rates, however, were much higher than their measured atmospheric disintegration rates, probably indicating either that the lifetime of the ^{38}Cl and ^{39}Cl atoms is too short for them to become quantitatively attached to filterable atmospheric aerosol particles or that their production rate is enhanced in the argon spheres by low energy radiation produced by the reaction 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 the latter effect.

On March 11, 1971, several spheres were filled with a total of 60 M^3 (STP) of argon and flown for 350 minutes at 15.3 km. The largest amount of argon that had been flown on any previous flight was 42 M^3 . These measurements permitted a significant improvement in the accuracy of cosmogenic radionuclide production rates; however, an attempt to measure ^{32}P (14.3 d) and ^{33}P (25 d) was unsuccessful even with very nearly quantitative chemical separations. Production rate measurement of ^{32}P and ^{33}P will require a long exposure of the argon-filled spheres at some high altitude location.

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.

A ^{85}Kr Field Monitoring System and Its Application to Maximum Air Dose Measurements of a Radioactive Gas J. D. Ludwick and P. W. Nickola

Our ^{85}Kr field monitoring system has been expanded by doubling the number of detector stations from 64 to 128 positions and by installing automatic cover plates on each detector which can be adapted to energy discriminate between two tracers.

An improved plume definition was sought by selective positioning of the added sensors although little, if any, improvement in sampling density was accomplished by the additional 64 stations. Monitoring stations were extended to the 1600 meter arc on the meteorological grid. Definition of the upper extent of typical plumes will be accomplished through measurements now available from sensors located up to 70 meters above ground elevation.

Modifications of the data collection center include the expansion of the programmer memory to identify information from 256 separate locations rather than the previous 64. In this way the instrumentation is

ready for any further expansion of the field locations beyond the existing 128. The real-time monitoring information collected on two 4096 address memory banks is also handled in a new manner. An electronic coupler was built to facilitate transfer of the 4096 information pieces to DEC-tape transports such as used in PDP-15 data processing computers.

There are several important advantages of this new system. The rapid information transfer ability of the coupler-tape system conserves real-time monitoring time, allowing continuous measurements of plume characteristics in time increments as small as 0.1 second. In addition, the computer compatibility feature of the new tape system allows data readout in a form designed to significantly reduce the man-hours previously needed for necessary information presentation.

The first problem investigated with the expanded ^{85}Kr monitoring system was one concerning the need for a more precise estimation of the maximum exposure near a nuclear installation in the event of a serious accident. For this purpose, a series

of 10 Ci ^{85}Kr sources were prepared in quartz vials. These vials, when crushed, simulate the rapid release of a dangerous radioactive cloud. The atmospheric transport of this puff-plume was then determined with the overall electronic system. In order to measure the consequences of accidental releases of this nature during unfavorable atmospheric conditions; releases were planned during relatively stagnant atmospheric conditions. To date, three releases have been made in the middle of the sampling grid, under rather light wind conditions, and during stable temperature-inversion characteristics. True stagnation conditions have not, as yet, been encountered; however, their seasonal appearance is anticipated and puff releases during these periods are planned.

Analytical Procedure for Measuring Copper in Biological Material

C. W. Thomas and L. A. Rancitelli

A technique has been developed to measure copper in biological material using neutron activation and electrodeposition. Samples are weighed under ultra-clean conditions in a laminar flow work station and placed in tared polyethylene irradiation containers which have been previously washed in doubly distilled nitric acid and rinsed with doubly distilled water. After weighing, the samples are freeze-dried to a constant weight, reweighed and the container heat sealed, and irradiated in a nuclear reactor together with standard solutions containing known

amounts of copper. The irradiated samples are transferred from the activation capsules to Erlenmeyer flasks containing 10 ml of fuming nitric acid, 50 mg of copper carrier and 20 mg of Na, K, and Br hold-back carriers. Each sample is digested for 2 hours, reduced to a final volume of 2 ml, transferred to a tared platinum evaporating dish, and diluted with 50 ml of water and 1 ml of concentrated sulfuric acid. The copper is electroplated onto the sides of the platinum evaporating dish by applying an electrical potential of 4 volts for one hour. After electrodeposition, the solution is discarded and the dish is washed several times with distilled water and finally with acetone. After drying under a heat lamp, the dish is reweighed to determine the chemical yield. The copper is dissolved with 2 ml of nitric acid, diluted to a constant volume and transferred to a standard counting geometry. The ^{64}Cu activity in the samples is measured by counting its annihilation radiation (a pair of 511 keV photons) with an anticoincidence shielded 9 in. diameter NaI(Tl) well crystal detector. The copper concentration in each sample is determined by direct comparison of the sample activity with that observed in known amounts of copper standards. The accuracy and precision of the technique was estimated to be 5% determined from replicate measurements of the copper content of standard reference materials (orchard leaves and beef liver) obtained from the National Bureau of Standards.

Analytical Procedure for Measurement of ^{214}Pb and ^{214}Bi in Rainwater

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

The short-lived daughters of radon, ^{214}Pb (26.8 min) and ^{214}Bi (19.7 min), are potentially useful tracers of precipitation scavenging processes because they are attached to the natural aerosols, they are present in easily measurable concentration in rainwater, and their half-lives are of the same order of magnitude as the time scale on which these processes occur. The major problem in using the radon daughters as tracers is the difficulty of rapidly separating them from large volumes of rainwater after short consecutive collection periods. A technique was recently developed to continuously separate ^{214}Pb and ^{214}Bi from rainwater at flow rates of up to 30 liters per minute and consecutive sampling periods of 5-10 minutes. Rainwater from a 3000 ft² polyethylene surface is

collected in a polyethylene 55 gallon drum and pumped through a fiberglass filter followed by an anion exchange bed (0.64 cm thick by 28 cm diameter of Dowex 1×8, Cl^- form, 200-400 mesh), and a cation exchange bed (0.64 cm thick by 28 cm diameter of Dowex 50WX8, H^+ form, 200-400 mesh). Bismuth-214 is removed at greater than 90 percent efficiency by the anion exchange bed. Bismuth-214 is measured immediately by counting the coincident photons emitted by the sample with a pair of 13 1/2 inch diameter by 6 inch thick NaI(Tl) detectors. No measurable amount of ^{214}Pb is removed by the anion exchange bed; however, greater than 90 percent of the ^{214}Pb is retained on the cation exchange bed which in turn does not retain ^{214}Bi . After the collection period the cation bed is set aside for about 1 hour to allow ingrowth of ^{214}Bi which is then measured using the detector system described and the ^{214}Pb concentration is calculated from this ingrowth.

Techniques Used to Stabilize Tracer Solution for Aerosol Generation

C. W. Thomas

Finely divided aerosols of several rare elements are generated by acetone-burner techniques on aircraft and serve as tracers in studying precipitation scavenging rates and mechanisms. Techniques were developed to put tracers of gold, iridium, ruthenium, tellurium and selenium either into solution or into a colloidal suspension in a solvent mixture that had excellent ignition and burning characteristics. A solution of acetone containing about 20 percent by volume of n-heptane was found to be compatible with almost any tracer that was moderately soluble in acetone and/or alcohol and had ignition and burning qualities that were compatible with a forced air aerosol generator. Laboratory and field studies were conducted and the following recipes were formulated and successfully demonstrated for both ground level and aircraft releases from acetone-burner generators of the types used in AgI releases.

Gold

175 grams $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$ was dissolved in 13 liters of acetone in a 50 liter polyethylene carboy. 3.2 liters of pyridine was added to prohibit reduction of gold by the metal surfaces of the generator. 6.5 liters of n-heptane was added. This solution was stable for two weeks after which the gold

solution was slowly reduced to the metal.

Iridium

32 grams of $\text{IrCl}_3 \cdot \text{XH}_2\text{O}$ was dissolved (with difficulty) in 10.5 liters of acetone by the following technique: the iridium chloride salt was stirred into a liter of acetone and the mixture was filtered. The filtrate was placed in a 50 liter polyethylene carboy. The residue was dried and fumed with concentrated hydrochloric acid and taken to just dryness. The residual salts were added to a liter of acetone and stirred and the mixture was filtered. The filtrate was added to the polyethylene carboy. This process was repeated until no appreciable amounts of residual salts remained after filtration. Additional acetone was added to make up to 16.5 liter volume and 4.5 liters of n-heptane was added. The solution was stable for weeks.

Ruthenium

150 grams of $\text{RuCl}_3 \cdot \text{XH}_2\text{O}$ was dissolved in acetone by the method used for iridium and made up to 16.5 liters with acetone. 4.5 liters of n-heptane was added. The resulting solution was colloidal but stable for one week after which slow coagulation occurred over a period of several weeks.

Tellurium

300 grams of TeCl_4 was dissolved in 16.5 liters of acetone and 4.5 liters of n-heptane was added. This solution was stable for weeks.

IN-CLOUD SCAVENGING

Several inert tracers were released into precipitating clouds near Quillayute, Washington and St. Louis, Missouri in order to determine precipitation scavenging efficiencies and aerosol and hydrometeor trajectories. Rain samples obtained from networks of rain collectors are being analyzed for the tracer materials by neutron activation techniques. Concentrations of the cosmogenic radionuclides ^{24}Na , ^{38}Cl , and ^{39}Cl were measured in large volume rain samples to determine scavenging efficiencies and elapsed times between scavenging and arrival of the precipitation at the ground. The concentrations of these cosmogenic radionuclides in argon and in air filter samples are also being measured to determine atmospheric production rates.

Precipitation Scavenging of Tracers Released into Rainstorms Near Quillayute, Washington

J. A. Young, W. E. Davis,
B. C. Scott, W. G. N. Slinn,
T. M. Tanner, C. W. Thomas,
J. M. Thorp and N. A. Wogman

Indium and lithium tracers were released from an aircraft at 5400 feet into precipitating clouds southeast of the Quillayute airport on March 11, 1971 in order to determine precipitation scavenging efficiencies and also aerosol and hydrometeor trajectories. Samples of rain were collected from the grid of 81 rain samplers, were frozen using dry ice and returned to the Richland laboratory for analysis.

Samples 15 through 38 and four reagent blanks were analyzed for lithium by atomic absorption spectroscopy. The concentrations were all below the detection limit

(0.01 ppm Li), so none of the remaining samples were analyzed for lithium.

Indium concentrations in the samples were determined by neutron activation analysis. Samples and standards were irradiated at the Washington State University 1 megawatt TRIGA III-fueled, water moderated, research reactor at a position with a neutron flux of approximately 8×10^{12} neutrons per cm^2 per second. Most of the samples contained measurable amounts of indium.

Large volume rain samples were collected at the Quillayute airport and at Rayonier's lumber camp, Camp Hoko, throughout the rainstorm for analysis for the cosmogenic radionuclides ^{24}Na (15 hr), ^{38}Cl (37 min), and ^{39}Cl (55 min). There did not appear to be any detectable systematic difference in the concentrations of the cosmogenic radionuclides between Quillayute and

Selenium

Selenium tetrachloride was not compatible with either acetone or n-heptane, both of which resulted in a rapid reduction to the metal. Alcohol was a suitable solvent for selenium, thus, 450 grams of SeCl_4 was dissolved in 4 liters of abso-

lute ethanol. This solution was metered simultaneously with n-heptane to the burner compartment of the generator. Since this required separate reservoirs for each solution the method has not been so suitable as others for aircraft release.

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.

Air Concentrations of ^{90}Sr and ^{55}Fe at Richland, Washington from 1963 to 1970

C. W. Thomas and J. C. Langford

The atmospheric concentrations of ^{90}Sr and ^{55}Fe were measured in ground level air samples collected at Richland, Washington from 1963 to 1970 as part of a program to define the rates of long-term stratospheric processes in the northern hemisphere. Seasonal variations in the concentrations of ^{90}Sr were similar to those of radionuclides of stratospheric origin, decreasing from 1963 to 1966 as a result of decay and deposition on the earth's surface. The Chinese thermonuclear tests at Lop Nor (45°N) beginning in 1967

maintained the surface air ^{90}Sr concentration at about the same level from 1967 to 1970.

The seasonal variations in the concentrations of ^{55}Fe were not as pronounced as those of radionuclides of stratospheric origin suggesting that a portion of the ^{55}Fe in the air at Richland, Washington came from the Hanford Project. This has previously been reported to be the case for ^{60}Co , ^{46}Sc , ^{65}Zn and ^{134}Cs . Discharge of reactor effluent water to the Columbia River causes buildup of radionuclides in river bottom materials and as the river level changes, some of the radionuclides in the dry areas become airborne.

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The Concentration of ^{55}Fe in Ground Level Air
at Richland, Washington from 1964-1968

Neg 720282-5

The Concentration of ^{90}Sr in Ground Level Air
at Richland, Washington from 1963-1970

The Determination of Air-Sea Exchange
and Oceanic Mixing Rates Using ^7Be
During the BOMEX Experiment

J. A. Young and W. B. Silker

The concentrations of ^7Be and other radionuclides were measured in air, rain, and seawater in a region east of Barbados, British West Indies, in the summers of 1968 and 1969 during the BOMEX experiment. The deposition of ^7Be on the sea surface by rainfall in August 1968 and May through July 1969 was only 13 and 26 percent, respectively, of that necessary to maintain the measured seawater inventories. The calculated deposition velocity of ^7Be on the sea surface by wet and dry deposition was 1.0 cm sec^{-1} . During May through June of 1969 the calculated flux of ^7Be across the sea surface averaged about $3.8 \times 10^{-2} \text{ atom cm}^{-2} \text{ sec}^{-1}$ and the inventories were increasing at a rate of $1.0 \times 10^{-2} \text{ atom cm}^{-2} \text{ sec}^{-1}$. Due to seasonal variations in the atmospheric ^7Be concentrations the ^7Be seawater inventories should vary seasonally with maximum inventories occurring around July and minimum inventories around February. The yearly average of the flux of ^7Be across the sea surface was calculated to be $1.6 \times 10^{-2} \text{ atom cm}^{-2} \text{ sec}^{-1}$. The vertical eddy diffusion coefficient, K_z , in the sea decreased from greater than $7 \text{ cm}^2 \text{ sec}^{-1}$ at the surface to 0.25 to $0.85 \text{ cm}^2 \text{ sec}^{-1}$ at 30 to 40 meters and then possibly increased somewhat from 40 to 100 meters. The ^7Be inventory in the sea varied rapidly due to the horizontal motion of seawater through the sampling region. The ^{103}Ru to ^{95}Zr ratio in the top 15 meters of the sea

indicated that large amounts of ^{103}Ru and ^{95}Zr from the French nuclear test series at 23°S beginning 7-7-68 entered the sampling region on 8-15-68 and also was present in June, but not in May 1969. The debris probably was carried into the sampling region by the Guiana Current which flows northwestward along the coast of South America.

Trace Element Characterization of
High Altitude Aerosol

T. M. Tanner and L. A. Rancitelli

In a continuing effort to measure the trace element content of atmospheric aerosols and to establish the origin, history and movements of various air masses, sampling missions were flown over the Pacific Ocean off the Oregon coast. Previous measurements of the trace element content of aerosols at altitudes of 3 to 15 km between Albuquerque, New Mexico ($34^\circ 05'\text{N}$, $106^\circ 45'\text{W}$) and Spokane, Washington ($45^\circ 36'\text{N}$, $117^\circ 38'\text{W}$) during 1967 and 1968 have been reported. It was shown that the concentrations of cobalt, iron and scandium could be explained by injection of material with the composition of the earth's crust into the atmosphere. It was also shown that the elements Sb, Ag, Zn, and probably Cr were far too high in concentration to have originated from the earth's crust. Sb and Zn were attributed to pollutant sources and Ag to extensive worldwide cloud seeding.

Measurement of the trace element profiles of the marine aerosol was essential to provide additional information on maritime air masses for our precipitation scavenging studies on the Washington coast. Missions were

flown during July, 1970 at altitudes of 3 to 12 km at a distance of 40 km off the Oregon coast (45°0'N, 124°34'W). The samples were taken and processed in the usual manner for analysis by neutron activation techniques. The aerosol samples taken at 9 and 12 km on July 27 and 28, 1970 have been analyzed and the results for eight elements, Ag, Br, Co, Cr, Fe, Sb, Sc and Zn, are presented in the accompanying table. Also presented for com-

parison are the concentrations of these elements at comparable altitudes in the samples taken over the continent. In the samples taken over the ocean, the concentrations were lower than over the continent. The trace element concentrations of these aerosols, when normalized to their iron concentration, show that with the exception of Ag and Br the ratios are nearly the same for both samples.

Comparison of the Concentrations of Several Elements in the "Maritime" Aerosol with "Terrestrial" Atmospheric Aerosols, Urban Ground Level Aerosol, Seawater and the Earth's Crust.
(All values normalized to Fe)

	Marine Aerosol(a)	Terr. Aerosol(b)	Sea Water	Urban Ground Level	Earth's Crust
Ag	0.26	0.035	0.08	0.041	0.001
Br	0.8	<0.004	1.9×10^4	0.05	0.00005
Co	0.00062	0.00092	0.1	0.0004	0.0005
Cr	0.0125	0.034	0.059	0.00006	0.002
Sb	0.0005	0.0014	0.097	0.004	0.000004
Sc	0.00022	0.00021	0.0012	0.0002	0.00044
Zn	0.225	0.21	3.2	0.2	0.0014

a. Average of two samples taken at 9 km and 12 km on 7-27-70 and 7-28-70, respectively.

b. Average of nine samples taken between 3 km and 15 km from August 1967 to October 1968.

Trace Element Concentrations Measured over the Pacific Ocean and Western U.S.
ng/scm of Air

Altitude, km	Volume of Air, 10 ³ scm	Ag	Br	Co	Cr	Fe	Sb	Sc	Zn
8.8 (Terrestrial) (10-28-68)	10.5	22±4	<1	0.022±0.008	2.7±1.5	71±12	0.29±0.05	0.008±0.002	8.7±1.4
9.1 (Maritime) (7-28-70)	12.7	2.1±0.3	1.80±0.03	0.003±0.001	0.05±0.05	4±1	0.002±0.001	0.0011±0.0001	1.1±0.1
11.4 (Terrestrial) (2-24-68)	3.55	0.03±0.02	<1	0.059±0.016	4±4	45±8	0.005±0.004	0.003±0.001	1.4±1.0
12.1 (Maritime) (7-27-70)	7.64	0.018±0.005	4.4±0.1	0.002±0.002	---	4±4	0.002±0.002	0.0007±0.0007	0.7±0.7

Comparison of Atmospheric Radio-
nuclide Concentrations at Neah Bay
and Richland, Washington

C. W. Thomas

The atmospheric concentrations of several radionuclides were measured continuously in ground level air from 1968 through 1970 at Richland, Washington (46°N, 118°W) and west of Neah Bay, Washington on the Makah Indian Reservation (49°N, 125°W) near the Pacific coast. Separating these two sites are the coastal and Cascade mountain ranges. Storm systems originating in the Aleutians pass over the Makah sampling site and then are orographically lifted several thousand feet before descending to the Richland sampling site.

At both Richland and Makah, large 350 cubic feet per minute air pumps are mounted several feet above the ground and draw air through membrane filters. The radionuclide concentrations on the filters are measured using large volume NaI(Tl) gamma-ray spectrometers. The concentrations

of ⁷Be, ⁹⁵Zr, ¹⁰⁶Ru, ¹³⁷Cs, and ¹⁴⁴Ce at the two sites show the seasonal variation that is characteristic of radionuclides of stratospheric origin. However, the concentrations are generally higher in the Richland samples, especially during the summer when the concentrations are at a maximum.

The lower average concentrations at the Makah site may be at least partially due to precipitation scavenging, since the rainfall rate is at least tenfold higher at that site. However, the difference in concentration between Makah and Richland is greatest in the summer when rainfall rates at both locations are relatively low, and is smallest in the winter when precipitation is heavy. It seems more likely that the high concentrations at Richland result primarily from vertical mixing associated with passage of air over the coastal and Cascade mountain ranges. Radionuclide concentrations are much higher at higher altitudes, so vertical mixing would result in higher

ground level concentrations. Supporting this explanation is the fact that ^{24}Na concentrations measured at

ground level at Richland were equal to the production rate at 7000 feet.

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Comparison of Radionuclide Concentrations in Air at Richland
(46°N, 117°W) and Makah (49°N, 125°W), Washington as a
Function of Time

Vertical Profiles of Radionuclide Concentrations in the Atmosphere

J. A. Young and N. A. Wogman

Vertical profiles of atmospheric radionuclide concentrations have been determined on a monthly basis since 1967 from air filter samples collected by RB-57 aircraft at 1.5 to 3.0 km altitude intervals from 0.3 km to an altitude of over 18 km. Most of the vertical profiles were measured either south of Spokane, Washington at 47°N, 117°W, or east of Barbados, British West Indies from 12 to 18°N and 54 to 59°W. The profiles at Barbados were taken during the summer of 1969 as part of the BOMEX project. The concentrations of cosmogenic radionuclides, nuclear weapons produced radionuclides, and radon and thoron daughters were determined using NaI(Tl), multidimensional gamma ray spectrometers and, beginning in late 1971, a dual, anticoincidence shielded, Ge(Li) gamma-ray spectrometer.

At Barbados the radionuclide concentrations generally did not increase systematically with altitude from ground level to 9 to 12 km, but then increased rapidly at higher altitudes. At higher latitudes in the northern hemisphere the concentrations generally increased continuously with increasing altitude at all altitudes. Apparently radionuclides which have been transferred from the stratosphere into the troposphere through the tropopause discontinuity at mid-latitudes have time to become fairly

well mixed vertically before they reach Barbados. A surprising feature of the profiles at all latitudes has been the fairly common occurrence of very pronounced concentration minima at altitudes from 6 to 9 km. Sometimes the concentrations at these altitudes are orders of magnitude lower than the concentrations at lower elevations. The only commonly measured radionuclide which does not show these concentration minima is cosmogenic ^{24}Na (15 hr). Na-24 is produced in the atmosphere at a relatively constant rate by spallation reactions of cosmic rays with atmospheric argon. Its half-life is short enough so that its atmospheric concentrations are relatively constant. Apparently, whatever process causes the concentration minima occurs long enough before sampling to allow ^{24}Na to grow back to nearly equilibrium. The presence of ^{24}Na at normal concentrations in the samples where minimum concentrations of the other radionuclides occur eliminates the possibility of sampling error as a cause of the minima. Most of the concentration minima probably result from precipitation scavenging, however the minima occur at altitudes above the most common altitudes of rain formation. It may be that the air is scavenged and then rises to 6 to 9 km before being sampled. The minima in the Barbados region may be due to air from the giant cumulonimbus of the intertropical convergence zone south of Barbados which reach altitudes of nearly 15 km. Another possibility is that the minima result

from scavenging by cirrus clouds. The cirrus ice particles may scavenge radionuclide bearing aerosol particles, either during or subsequent to nucleation and then settle, leaving a region of low radionuclide concentration behind.

In the past, efforts have been made to calculate back trajectories of the air containing minimum radionuclide concentrations in an attempt to determine the cause of the minima. However, the lack of meteorological stations on the ocean upwind of the location of the profile measurement has thwarted these efforts. Therefore, vertical profiles have been measured

in 1971 farther inland over the continental United States. A profile measured on February 24, 1971 near Albuquerque at 35°N, 106°W showed concentration minima at 9.1 km, and a profile measured on November 16, 1971 near Denver at 39°N, 103°W showed a concentration minimum at 6.1 km. In both cases the ^{24}Na concentration was only slightly below normal. The concentration of a few radionuclides on November 16 are plotted as a function of altitude in the figure. Back trajectories are being calculated for both cases in an attempt to identify the cause of the minima.

Radionuclide Disintegration Rates on November 16, 1971
at 39°N, 103°W

Atmospheric Fallout During 1971

C. W. Thomas and J. A. Young

The atmospheric concentrations of radionuclides in ground level air were measured continuously during 1971 at Point Barrow, Alaska (71°N) and Richland, Washington (46°N) by filtering large volumes of air through membrane filters followed by gamma-ray spectrometric analysis. The concentration of nuclear-weapons-produced radionuclides, including fission products and neutron activation products, increased steadily from 1967 through 1971 because of the high yield thermonuclear tests conducted by the Chinese at Lop Nor (45°N) during this period with a total yield of over ten megatons. During 1971 essentially all of the nuclear-weapons-produced radionuclides present in northern hemispheric air originated from these Chinese tests. The concentration of ^{88}Y (107 day half-life) increased 100-

fold over 1968 peak levels and during 1971 reached peak concentration levels comparable with peak concentrations of 1962-1963.

The Chinese nuclear test of 11-19-71 was detected in low concentrations in ground level air at Richland, Washington five days after detonation. During 1970 the nuclear reactors, which are a part of the Richland Atomic Energy complex and use Columbia River water as a straight through coolant, were shut down. The effect of this shutdown was noted by a decrease in air concentration of several radionuclides that were known to be associated with the Richland operations. Scandium-46 concentrations in air were down by two orders of magnitude, ^{60}Co was down one order of magnitude, and ^{124}Sb is now below detection levels. Also affected was ^{65}Zn air concentration which now shows an excellent seasonal variation and is down by an order of magnitude.

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 in Pacific Salmon, 1965-1970

C. E. Jenkins and J. C. Langford

Iron-55 and stable iron measurements are being made to help characterize the ^{55}Fe uptake and movement through the marine biosphere. Particular attention is being focused on salmon and their food chain since salmon as a food source are responsible for the highest ^{55}Fe concentrations observed in man. Results of this study show the following: ^{55}Fe concentrations are more than an order of magnitude higher in salmon from the Bering Sea than in those from waters off the contiguous United States; ^{55}Fe in salmon has an apparent effective half-life of 9 to 10 months; ^{55}Fe entering the ocean from the atmosphere is rapidly assimilated by the oceanic biomass with little apparent recycling; ^{55}Fe concentrations in the oceanic organisms are thus declining about 2 times faster than in the terrestrial herbivores; Kotzebue chum salmon are 3- to 10-fold higher in ^{55}Fe than Alaskan caribou, and 300 to

1000 times higher than cattle from Columbia Basin, Washington; salmon-eating Eskimos have 20- and 100-fold higher ^{55}Fe body burdens than those of caribou-eating Alaskan Eskimos and Richland, Washington cattle-eating residents, respectively.

This study indicates that high ^{55}Fe concentration in marine organisms from Alaskan waters is not the result of a high initial fallout from Russian thermonuclear testing and recycling, as has been suggested by some authors. The phenomenon of increasingly higher radionuclide concentrations in salmon from more northerly waters is not confined to ^{55}Fe . Both ^{54}Mn and ^{60}Co appear to behave similarly. Cesium-137 levels in seawater confirm land measurements of fallout which indicate that fallout is less in the northern regions than at the mid-latitudes. Since fallout rates are indeed less at northern latitudes, while concentrations in ocean organisms are higher, this suggests that a very efficient mechanism exists in northern oceanic waters for concentrating

fallout elements. This has extremely important implications with regard to all anthropogenic toxic trace elements and suggests that a most critical area in the marine biosphere is in the northern regions of our oceans.

The cause of this phenomenon is not yet understood; however, it is

being seriously investigated. The implications are, of course, that not only radioelements but many toxic trace elements that are currently entering the oceans from the atmosphere will be most highly concentrated in the marine biosphere in the northern regions of the ocean.

Neg 711947-2

Total Body ^{55}Fe Burden in Adult Alaskan Eskimos
and Richland, Washington Residents

⁵⁵Fe Concentrations in Pacific Salmon

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 Performance and Application of a Versatile Ge(Li)-NaI(Tl) Coincidence-Anticoincidence Gamma-Ray Spectrometer for Environmental and Biological Problems

J. A. Cooper and R. W. Perkins

A high-sensitivity gamma-ray spectrometer incorporating a high-performance Ge(Li) detector and a NaI(Tl) well crystal has been developed, which provides improved sensitivities for coincident and noncoincident gamma-ray emitting radionuclides commonly found in environmental and biological samples. As an anticoincidence spectrometer it provides the highest peak-to-Compton edge ratio ever observed, 780:1, over 3 times greater than any other known system. Interference from such coincident gamma-ray emitting radionuclides as ^{60}Co is reduced by 80-fold in the anticoincidence spectrum. This high efficiency for removing Compton interference, the 10-fold suppression in natural background, and the high absolute efficiency of the Ge(Li) detector (27% for a 122 keV point source at

the cap), provides the most sensitive system yet described for measuring radionuclides such as ^{137}Cs in environmental samples.

Normal plastic shielded anti-coincidence spectrometers have not been able to significantly improve the sensitivity for the measurement of coincidence gamma-ray emitting radionuclides. However, because of the resolving capabilities and the high efficiency of the NaI(Tl) well crystal used in this spectrometer, it is possible to select specific-energy coincidence events occurring in the NaI(Tl) and Ge(Li) detectors with about the same counting efficiency as with a normal Ge(Li) detector and to obtain tremendous improvements in the sensitivity for the measurement of coincident gamma-ray emitting radionuclides. Background interferences have been reduced by as much as 400 times. In the analysis of caribou liver samples, the peak to background ratio for the 434 keV peak of $^{108\text{m}}\text{Ag}$ was improved by about 350-fold. This improvement was in a coincidence spectrum which was gated on the

1338 keV ^{108m}Ag sum peak in the NaI(Tl) detector. The coincidence spectra for all of the coincident gamma-ray emitting radionuclides can be accumulated simultaneously along with the anticoincidence spectrum through the use of three coincidence windows. By using all of the time and energy information provided by the Ge(Li) and NaI(Tl) detectors, significant improvements in sensitivity for both the coincident and noncoincident gamma-ray emitting radionuclides can be ob-

tained. Sensitivities for 1000 minute counting intervals range from a few tenths to one dpm for most of the radionuclides measured in typical environmental and biological samples. These sensitivities are based on a maximum source size of 5 cm diameter by 1 cm thick. As the sources become substantially larger, as in the case with some environmental samples, the sensitivity rapidly decreases and the large multidimensional gamma-ray spectrometers become the instrument of choice.

Neg 711600-11

The Sensitivity of Instrumental Analysis of Complex Mixtures of Radionuclides With Ge(Li) Detectors is Greatly Improved by Removing the Compton Interference with the NaI(Tl) Shield Which Provides an 18-fold Reduction of the ^{137}Cs Compton Edge (Almost Twice That of Previously Reported Systems) to Provide a Peak-to-Compton Minimum of 915:1 and a Peak-to-Compton Edge Ratio of 780:1 Which is More Than Three Times That Obtained With Previously Reported Systems.

Neg 711984-5

The Ge(Li)-NaI(Tl) Spectrometer not Only Improves the Sensitivity for Noncoincidence Gamma-ray Emitting Radionuclides but Also Provides Improved Sensitivities for Such Coincidence Gamma-ray Emitting Radionuclides as ^{108m}Ag Where the Compton Interferences can be Reduced by as Much as 400 Times to Yield About a 350-Fold Improvement in the Peak to Background Ratio and Sensitivities of a Few Tenths of a dpm.

Natural Background Radiation in NaI(Tl) and Ge(Li) Gamma-Ray Spectrometers

N. A. Wogman and J. A. Cooper

Since natural background plays an important role in the analysis sensitivity of gamma-ray spectrometers, a study was made to define the sources of background and to discover methods to reduce their effect in gamma-ray

spectrometer systems. The systems used in the investigation were of three general types: single Ge(Li) or NaI(Tl) crystals; anticoincidence-shielded Ge(Li) or NaI(Tl) crystals; and, anticoincidence-shielded multi-dimensional NaI(Tl) and Ge(Li) gamma-ray spectrometers.

In the energy range from 0 to 4 MeV, the primary contributions to

the background in non-hydrogen containing systems are the gamma-rays emitted by members of the ^{238}U and ^{232}Th decay chains and ^{40}K . Alpha particles from the decay of ^{238}U and ^{232}Th also produce background peaks from 3 to 8 MeV. These alpha particles were observed in 10,000 minute backgrounds in NaI(Tl) crystal systems, but not in the Ge(Li) spectrometers. The cosmic-ray induced background results from the passage of primary or secondary particles through the detector and from the reaction of these particles with surrounding materials producing neutrons which may interact with the system materials to produce radioactive contaminants. In those systems containing hydrogen; e.g. plastic phosphors, a very apparent background photopeak results from the prompt gamma ray from cosmic-ray neutron interactions with hydrogen [$^1\text{H}_1$ ($^1\text{n}_0$, 2.225 MeV γ) $^2\text{H}_1$]. In addition, these neutrons interact with I of the NaI(Tl) detector and with the Pb or Fe construction materials to produce prompt gamma-rays of 6.8, 7.4, and 7.6 MeV, respectively. Cosmic-ray particles de-

posit energy in the NaI(Tl) and Ge(Li) crystals directly. For example, the most probable energy loss of a 1 GeV muon in NaI is 1.47 MeV cm^2/g . This process creates a high energy peak and an energy continuum throughout the range of 0-10 MeV.

A threefold background reduction of the neutron induced peak was achieved by lining the shield with four inches of borated (5% by weight) paraffin to thermalize and capture all neutrons produced in the external shields. A concurrent background reduction of 15% occurs over the energy range of 0 to 3 MeV. The internal neutron component was not eliminated. The lowest overall backgrounds were obtained by a combination of methods to reduce the individual background sources: (1) eliminating external natural radiation by use of uncontaminated construction materials, (2) borated paraffin shielding to moderate and capture externally-produced neutrons, and (3) installation of the instrument deep underground where the massive overburden attenuates the major portion of the cosmic ray muons.

Natural Radioactivity in NaI(Tl) and Ge(Li) Gamma-Ray
Spectrometer Construction Material Often Precludes Low-
Background Utilization and Its Variance from Sample to
Sample Requires Analysis for ^{232}Th , ^{238}U , and ^{40}K Prior to Use

Spectrometer Assembly Material	dpm/g per sample		
	^{232}Th	^{238}U	^{40}K
Al (25)	0.12	<0.07	1
Al (6061)	0.42	0.04	<0.05
Al (1100)	0.24	<0.017	<0.06
Al (1100)	0.08	<0.026	<0.11
Al (3003)	0.10	<0.026	0.56
Al Sheet	<0.08	<0.1	<0.4
Al "	<0.1	<0.2	<0.5
Al "	0.9	<0.007	<0.09
Al "	0.3	<0.007	<0.09
Al ₂ O ₃ (Reflector)	0.11	<0.07	<0.3
CdF ₂ (Light pipe)	<0.003	<0.003	<0.01
Cu (Sheet)	<0.005	<0.006	<0.2
Cu (Rod)	<0.02	<0.03	<0.09
Ge Ingot	<0.02	<0.02	<0.05
Ge Ingot	<0.02	<0.02	<0.04
Ge Cylinder	<0.02	<0.02	<0.04
Ge "	<0.01	<0.01	<0.02
Fe Plate	<0.003	<0.005	<0.01
Fe 304 Stainless	<0.006	0.007	0.06
Fe 309-L Stainless	<0.005	<0.005	<0.02
Mg Rod	0.06	<0.04	0.1
Mg Ingot	<0.01	<0.002	<0.02
MgO Reflector	0.088	0.14	<0.23
MgO Reflector	<0.05	<0.05	<0.2
Pb Shavings	<0.008	<0.01	<0.03
Pyrex Window	0.45	0.27	3.8
Quartz "	<0.018	<0.018	<0.07
Quartz "	<0.018	<0.018	<0.07
Polyvinyl Chloride foam	1.0	0.9	1.6
" " "	0.19	0.4	0.36
" " "	<0.2	0.4	<0.7
Neoprene	<0.008	<0.01	0.36
Rubber	0.12	1.2	1.8
"	0.12	1.0	2.0
Mattress Foam-U.S. Rubber	1.9	15	2470
Apiezon Q	4.5	4.5	2.7
Electrical Tape - 3M	<0.04	<0.06	<0.1
Cement - Portland	0.25	1.3	4.5
Molecular Sieve	4.4	3.0	9

Improved X-Ray Fluorescence Analysis
of Environmental and Biological
Samples Using a High-Intensity Radio-
isotope Excitation

J. A. Cooper

The elucidation of the role of beneficial and detrimental trace elements in environmental and biological systems is basic to our understanding of their environmental balance. Because studies of behavior of trace elements in environmental and biological systems often require the analysis of a large number of samples, many of these studies have been conducted only in a superficial manner because of the lack of sensitive, rapid, and practical techniques of analysis.

Improved X-ray fluorescence techniques which employ radioisotopic sources and which should provide sensitivities comparable to those from machine excitation are being developed for environmental and biological specimens. A high-intensity radioactive source of photons is used to excite the elemental X-rays which are analyzed with a Si(Li) diode. The radioactive sources (^{55}Fe , $^{109\text{m}}\text{Cd}$ and ^{241}Am) are about ten times more intense than previously available sources and the holders are doubly collimated to minimize nonsample scat-

tering. A large vacuum chamber is also used to eliminate the air scattering, one of the major sources of interference remaining for thin samples. The technique is capable of providing sensitivities in the ppm range for most environmental matrices.

A large variety of environmental samples have been analyzed for trace elements. Sixteen elements (S, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ge, Br, Sr, Zr and Pb), many of which (S, Ca, Ti, V, Ge, Sr and Pb) are not readily analyzable by other techniques such as neutron activation analysis, are measured in many types of samples in just 30 minutes using 100 mCi $^{109\text{m}}\text{Cd}$ source. Samples of suspended particulates from a uranium mine and animal test chambers have been analyzed for over a dozen elements (Si, K, Sc, Ti, V, Mn, Fe, Cu, Zn, Pb, Se, U, Sr, Y and Zr) many of which would have been quite difficult by neutron activation analysis because of the large interference from U fission products. Other samples such as sea flounder muscle tissue, seawater, urine, finger nails and geological samples have also been examined. Techniques for preparation of thin environmental and biological samples, for obtaining more precise quantitative results, and for increased automation of the analysis are being developed.

Neg 715596-1

Elemental Content of Biological and Environmental Samples Such as Air Filters are Measured Rapidly and Nondestructively with X-ray Fluorescent Techniques and High-Intensity Radioactive Sources. This Technique Provides the Capability of Studying Large Numbers of Samples Required in Many Environmental and Biological Investigations

X-ray Fluorescence Determination of the Zinc to Copper Ratios in Fingernails for the Identification of Cystic Fibrosis

J. A. Cooper, C. E. Jenkins,
 J. C. Langford and N. A. Wogman

Cystic Fibrosis, which has an incidence of approximately one per thousand births, must be diagnosed early in order that permanent irreversible damage to the lungs and other body organs be prevented. Early detection permits the initiation of treatment which helps keep the effects of this disease in check.

The analysis of fingernails or sweat for sodium has been the method most frequently suggested as a possible means for mass screening of children for the disease; however, the reliability of this method is not very satisfactory.

A rapid and inexpensive technique for the multielement analysis of trace elements in nail clippings has been developed, based on excitation of the characteristic X-rays of the elements in the nail with a high-intensity radioisotopic source and their analysis using a Si(Li) diode. The sensitivities for many elements

are in the parts-per-million (ppm) range and the procedure could be easily automated, making it a very useful tool for scanning the trace element distribution in a large number of individuals. The elements S, Cl, K, Ca, Fe, Co, Ni, Cu, Zn, Pb, Br and Sr have been measured in fingernails from about 6 confirmed cystic fibrosis cases and 50 normal individuals. The Cu content of fingernails from individuals with cystic fibrosis is usually higher than normal while the Zn content is usually lower than normal. Thus, the ratio may be a sensitive indicator and free from many of the normal errors associated with an absolute concentration determination.

Preliminary results indicate the Zn:Cu ratio of normal individuals is about 7:1 while the ratio for those with cystic fibrosis is about 3:1. Unfortunately, the spread in the distribution is quite high, which may be due to physical differences in the nail clippings analyzed or the small number of samples used to define the distribution. A technique to provide sample uniformity is being developed, which consists of a quick dissolution of a weighed nail sample followed by evaporation on a thin Mylar film. This should allow precise determination of the Zn:Cu ratio under controllable conditions. A large number of nail clippings from both normal and cystic fibrosis individuals have been obtained from the University of Washington and will be used to accurately define the variation in the Zn:Cu ratio in the two groups.

Transmission Properties of Specific Scintillator Reflective Coatings

R. L. Brodzinski

Four 40-inch diameter by 12-inch thick sections of plastic scintillator, which were to make up an anti-coincidence shield, were found to be inefficient in transmitting light to their phototubes. Test sections of these ingots demonstrated satisfactory scintillation properties; therefore, it was recognized that the difficulty was related to the reflective coatings applied to the phosphors' external surface. Tests were conducted on a small core of scintillator material (Pilot Y) with the criterion for good performance being a high efficiency for detection of ^{137}Cs gamma rays. The effect of reflective coating thickness on photon detection efficiency was evaluated for three different formulations supplied by Pilot Chemical Company. Maximum photon counting efficiency was observed for coatings between 0.0025 and 0.0030 inch thick. With optimum coating thicknesses it was found that all coating formulations yielded approximately the same efficiency. On studying light-tight materials such as aluminum foil, aluminized Mylar, and black plastic, it was found that they greatly affected the transmission losses from the reflective coatings. Though the transmission losses were small in all cases, there were two to threefold differences in the light output.

Optically coupling the reflective coating to the light-tight wrapping was found to enhance the transmission of light through the coating. Therefore, optical coupling should be avoided, and the light seal material should be wrapped over a clean dry surface. The reflective properties of aluminum foil and aluminized Mylar are very low compared to those of the spray-on reflective coatings and should not be used as a primary reflector.

Computer System for Radionuclide Analysis

J. R. Kosorok

Standard procedures in instrumental neutron activation analysis require the simultaneous irradiation of standards and samples. After irradiation, the gamma-ray spectra of the radionuclides in the samples and standards are acquired and compared to determine element content. Quantitative analysis requires the spectral evaluation together with the weights of standards and samples, radionuclide half-lives, shape and location of the characteristic photo-peaks of the radionuclides, irradiation and counting times and counting intervals. When large numbers of routine samples are involved, as much of the analysis procedure as possible should be automated to reduce the time required to perform the analysis. A small computer system has been programmed for routine use in these evaluations.

In using the system, the operator directs the computer system to com-

pose and maintain a library of constant, characteristic data for a maximum of 40 radionuclides; acquire, display and store gamma-ray spectra; and perform the necessary computations to determine element concentrations. The computer automatically collects spectra and calculates element concentrations with little direction from the operator. When not acquiring spectra or processing acquired data, the system can be used by the operator to initialize, expand or modify the radionuclide constants library. A researcher may start with a library containing data on a few isotopes and expand it as his samples become more complex.

Any pulse-height-to-digital-converter which generates a live/clock pulse in channel zero can be interfaced to the computer system to acquire spectra for time intervals set by the operator. In addition to measuring the elapsed counting interval, the system has a real-time clock and stores the time and date at which a spectrum was collected. The time of count is used internally to correct for radionuclide decay between the time of irradiation and time at which the gamma-ray spectrum was acquired. A scope display of the spectrum may be requested during or after data acquisition. Live time is not appreciably affected by the display, because the display has direct access to the computer memory rather than being entirely program controlled. The spectrum

for a particular standard or sample along with enough data to identify it for further analysis may be listed on a printer for the operator's study as well as stored on magnetic tape for automatic processing.

A line in the report printed by the system identifies each element, the decay corrected gamma-ray count of its activation product, and its concentration in the sample. The computer system was developed by integrating commercially available hardware and software with interfaces and software developed at Battelle-Northwest. The hardware for the system includes a Digital Equipment Corporation PDP-15/30 computer, a pulse-height-converter, a display unit, and a fast printer. All programs performing operator requested functions operate under control of the monitor provided by the computer manufacturer. Programs communicating between the monitor and the interfaces for the pulse-height-converter, display unit and fast printer were developed in Battelle-Northwest.

The computer system for instrumental neutron activation analysis described here has been routinely used for determining trace amounts of elements in environmental studies. The software design has allowed modification and expansion to fulfill the changing needs of research and has provided a flexible and powerful tool for scientists.

Multielement Analysis of Biological Reference Materials

L. A. Rancitelli

The elemental content of biological materials, particularly the trace constituents, has received a great deal of interest in the last few years. This recent interest has been spawned by the realization that inorganic pollutants such as mercury have permeated the biosphere and invaded the food chain of man. Additional interest in the trace inorganic constituents of the biosphere has been generated in studies of the radiologically important fallout and reactor associated radionuclides in the environment. Recognizing the need for biological standards both for the comparison of analytical techniques and for providing inter-laboratory comparisons, the Division of Biology and Medicine of the Atomic Energy Commission supported the National Bureau of Standards efforts to prepare and certify botanical and animal tissue reference materials. This effort has included their certification of a standard orchard leaf, SRM 1571. The analysis of the orchard leaf and two other botanical materials, under consideration as possible botanical standards, tomato leaves and citrus leaves, was conducted at our laboratory using a highly sensitive and precise technique of instrumental neutron activation developed at the Pacific Northwest Laboratories.

Briefly, the method consisted of irradiating 100 mg samples of freeze-dried material in a Hanford production reactor to an integrated neutron exposure of 10^{18} n/cm². A gamma-ray analysis of the activated samples, after appropriate decay intervals, was made with both a Ge(Li) detector system and an anticoincidence shielded multidimensional analyzer to measure the neutron-induced activities ²⁴Na, ⁴²K, ⁴⁶Sc, ⁵¹Cr, ⁵⁹Fe, ⁶⁰Co, ⁶⁵Zn, ⁷⁵Se, ⁷⁶As, ⁸²Br, ⁸⁶Rb, ^{110m}Ag, ¹²⁴Sb, ¹³⁴Cs, ¹⁴⁰La and ²⁰³Hg. A comparison of these activities with those induced in known amounts of each element irradiated with the samples permitted the determination of 16 elemental concentrations in each sample.

Seven samples of orchard leaves, and 4 samples each of tomato leaves and citrus leaves were analyzed by neutron activation analysis. The results are presented in the table

together with the mean concentration and standard deviation. In the case of orchard leaves, the agreement between the results of this work and those elements certified by NBS, sodium, potassium, arsenic, rubidium, iron, mercury, selenium and zinc, are generally good. In addition to those elements certified by NBS, we also report in the table the concentrations of bromine, chromium, cobalt, silver antimony, and cesium. The utility of the citrus leaves as a standard is doubtful due to its gross inhomogeneity as indicated by the high variability in the iron and cobalt content. The tomato leaves have a uniform trace element content much like that of the orchard leaves. However, it is questionable if the differences in elemental composition between the certified orchard leaves and the tomato leaves justifies the adoption of the latter as a second botanical standard.

Elemental Content of Biological Materials
Determined by Neutron Activation Analysis ($\mu\text{g/g}$)

<u>Element</u>	<u>Orchard Leaves**</u>	<u>Citrus Leaves</u>	<u>Tomato Leaves</u>
K	13,700 $\pm$ 600	20,300 $\pm$ 1400	44,600 $\pm$ 2000
Na	78 $\pm$ 3	190 $\pm$ 14	540 $\pm$ 12
Fe	300 $\pm$ 45	121 $\pm$ 52	460 $\pm$ 36
Zn	27.2 $\pm$ 2.4	29.8 $\pm$ 1.8	62 $\pm$ 1
Rb	10.3 $\pm$ 0.6	4.3 $\pm$ 0.4	14.1 $\pm$ 1.2
Br	9.2 $\pm$ 0.2	9.9 $\pm$ 0.6	24.4 $\pm$ 1.1
As	15.4 $\pm$ 0.2	4.6 $\pm$ 0.2	---
La	2.1 $\pm$ 0.1	0.37 $\pm$ 0.08	0.71 $\pm$ 0.08
Co	0.14 $\pm$ 0.01	0.021 $\pm$ 0.007	0.42 $\pm$ 0.02
Sb	2.7 $\pm$ 0.3	0.042 $\pm$ 0.005	0.029 $\pm$ 0.004
Hg	0.18 $\pm$ 0.02	0.14 $\pm$ 0.02	0.09 $\pm$ 0.02
Sc	0.044 $\pm$ 0.003	0.0067 $\pm$ 0.0006	0.080 $\pm$ 0.005
Se*	0.14 $\pm$ 0.02	0.06 $\pm$ 0.02	<0.1
Ag	<0.02	0.006 $\pm$ 0.002	0.019 $\pm$ 0.002
Cs	0.029 $\pm$ 0.002	0.093 $\pm$ 0.005	0.050 $\pm$ 0.003
Cr*	1.8 $\pm$ 0.4	0.46 $\pm$ 0.05	1.8 $\pm$ 0.1
No. of Analyses	7	4	4

* Tentative Values

** National Bureau of Standards, SRM-1571

Characterization of Columbia River
Basalt Flows From Major, Minor and
Trace Element Considerations*

L. A. Rancitelli

The safe storage of radioactive wastes at Hanford requires a comprehensive knowledge of the local geological structure. For example, faults in the bedrock as well as contact zones between geological strata can be potential avenues for the transport of liquid wastes into the ground water. The bedrock underlying the Hanford project is composed of more than 20 independent basalt flows ranging in thickness from a few feet to several hundred feet. Although these flows were deposited over a long period of time, they are very similar in their major element content, petrology and the physical properties usually employed to characterize them. These similarities make it very difficult to identify or recognize an individual basalt flow over distances of a few miles, an important point in understanding the geological characteristics of the region. One technique which has not been extensively investigated in the study of the local stratigraphy has been the trace element content of the Columbia River Basalts. In an effort to characterize individual basalt flows from the elemental composition, a multielement technique

of comparative instrumental neutron activation analysis capable of simultaneously measuring 17 elements with a high degree of sensitivity and precision was developed to support the AEC's waste storage programs. This approach has proved highly successful in characterizing the local stratigraphy of the Columbia River Basalts using samples taken from test wells.

Briefly, the technique involves a 10 second irradiation of a 100 mg basalt sample together with appropriate standards at a neutron flux of 10^{13} n/cm²·sec. After a 2 minute decay interval to reduce the interference from ²⁸Al, the sample was counted on a Ge(Li) spectrometer for 2 minutes. This irradiation permits the simultaneous measurement of Al, Mg, Ti, Na, V, and Mn. The sample and appropriate standards are then re-irradiated to an integral neutron flux of 10^{17} n/cm², decayed for 5 days and 20 days and counted again on a Ge(Li) spectrometer for periods of 10 and 100 minutes, respectively. The 10 minute count permits the measurement of La, Sm and Na, while the later 100 minute measurement provides the concentrations of Fe, Co, Sc, Eu, Th, Cr, Hf, Ta, and Tb. It will also be possible to measure Ca from the ⁴⁷Sc activity; however, the necessary calibrations are not yet complete. The technique provides a high degree of precision, on the order of 3% except when the detection limit of an element is approached, as demonstrated by 18 analyses of the U. S. Geological Reference Material BCR-1. This high precision is of paramount

* The technology development for this study and for the analysis of soils and sediments was supported by DBM. The stratigraphy study is not supported by DBM.

importance to the study since the concentration of some elements varies by only a few percent between basalt flows. As an example, the Sc content ranged from 36 to 47 ppm in the Atlantic Richfield Hanford Company test well DDH-3 from depths of 1680 feet to 2700 feet. However, other elements show significant differences between adjacent flows which have been successfully employed to characterize a flow uniquely. For example, Cr was noted to vary by 2 orders of magnitude between flows while varying only 10% within a flow, while Eu, La, Sm, Tb, and Ta varied two-to four-fold. A complete major, minor, and trace element characterization of the Columbia River Basalt flows promises not only to provide the necessary information to uniquely characterize each flow but may also provide valuable geochemical evidence bearing on the conditions which existed at the time of their formation.

Trace Element Content of Aquatic Organisms from the Columbia River

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

Stable element concentrations in the environment are an important parameter in our understanding of the transport and distribution of radionuclides throughout an ecological system. The stable element counterparts of radionuclides not only serve as carriers and diluents but in some cases such as Co, Fe, and Zn participate in biologically essential functions within an organism. Other elements such as As, Cd, and Hg, which

are known to be toxic to biological systems, can produce detrimental effects in the environment which can be serious. Thus, it is important to establish the present levels of the toxic heavy metals, biologically essential elements and radiologically important elements in the environment. In an effort to accomplish this end for the Columbia River system, an extensive study of 17 elements in seven Columbia River organisms has been made by instrumental neutron activation analysis. Dorsal muscle, liver and kidney samples from the 6 fish, chiselmouth, carp, squaw fish, coarse and fine scale suckers, white fish, and sturgeon, as well as caddis fly larvae were analyzed for the following elements: K, Na, Ag, As, Au, Br, Cd, Co, Cr, Cs, Fe, Hg, Rb, Sb, Sc, Se and Zn.

As was the case for marine organisms, silver was found primarily in the liver of the Columbia River fish. Cadmium levels of 1 to 20 ppm were found in the kidney, while mercury levels as high as 6 ppm in the kidney of a white fish were observed. Since the sturgeon has a much longer life span than other fish, a more extensive survey was performed which included heart muscle, notochord, gonads, pectoral ray and cartilage in addition to the dorsal muscle, liver and kidney. An arsenic concentration of 140 ppm (dry basis) was found in the gonads. The mercury content of all sturgeon samples was below 0.2 ppm on a fresh weight basis.

The caddis fly larvae had much higher levels of Fe (2000 ppm),

Sb (0.11 ppm) and Sc (0.5 ppm) than is normally encountered in biological tissue, probably reflecting the fact that a significant amount of inorganic detrital material was present in the organism.

Radionuclide Distribution in
Olympic National Park, Washington

C. E. Jenkins, N. A. Wogman and
H. G. Rieck

A survey of the radionuclide distribution in precipitation, river water, river sediments, soils, plants, and animals was conducted in the Olympic National Park of Northwestern Washington which has an annual precipitation ranging from 60 to 200 inches in less than 50 miles distance. Because of the high precipitation regime and geographic location, the total fallout radionuclide concentrations per unit area are relatively high. These relatively high concentrations permit the tracing of many of the less abundant and shorter-lived radionuclides through the biosphere.

Thirty-one cosmogenic, naturally occurring, and fallout radioelements were measured in air, rainwater, stream water, sediments, flora, and fauna samples to establish concentration levels and provide data on the relative behavior and fate of radioelements in a forest ecosystem. It was found that the total fallout concentrations per unit area in the Olympic National Forest were the highest observed for any non-nuclear

testing area in the world and generally were some three- to four-fold higher than for the Alaskan Arctic region. However, the radionuclide concentrations in an Olympic elk and deer appear 1.2- and 4-fold lower, respectively, than for Arctic Alaskan caribou. Olympic Peninsula residents who consume local produce and wild game have slightly increased levels of ^{137}Cs over Richland, Washington residents, whose levels are nearly 50- to 100-fold less than in caribou eating Alaskan Exkimos. Two distinct seasonal patterns emerge from Olympic flora fallout concentrations. The first pattern, which is typical of most Olympic flora, shows seasonal variations which are somewhat greater than seasonal variations in air concentrations. The second pattern, which is typical of slow growing mosses and lichens, shows less variation than fallout concentrations in the atmosphere.

A rough approximation of the radionuclide concentration factors from the troposphere through this forest biome are: air to rainwater, 10^6 to 10^7 ; rainwater to flora, 10^2 to 10^3 ; and flora to fauna, 10 to 10^2 . The input of radionuclides to the forest biome is much greater than loss from the system by runoff which indicates a net accumulation. This indicates that pollutant trace elements would also be accumulated. In view of present atmospheric levels of many hazardous materials, detailed studies of the biogeochemical behavior of these materials would be desirable.

Radionuclide Concentrations for Air and Rainwater
1966-1967 Olympic Peninsula, d/min/liter

Sample Date Location Units			Air ⁽¹⁾ 11/66 Richland 108ℓ	Air 3/67 Richland 108ℓ	Air 1/68 Richland 108ℓ	Air 12/67 ONP 108ℓ	Rainwater 11/15/66 ONP 10 ³ ℓ	Rainwater 3/21/67 ONP 10 ³ ℓ	Rainwater 1/11/68 ONP 10 ³ ℓ
Radionuclide	t _{1/2}	Source ⁽²⁾							
⁷ Be	53d	C	1050	1360	1200	1280	26,300	56,700	66,500
²² Na	2.6y	C	0.068	0.28	0.127	0.102	----	17.2	18.6
⁴⁰ K	1×10 ⁹ yr	N	----	----	----	----	----	565	----
²⁰⁸ Ti(²³² Th)	1.39×10 ¹⁰ yr	N	0.096	0.74	0.26	0.13	4.0	----	----
²¹⁴ Bi(²²⁶ Ra)	1620yr	N	0.19	0.48	0.93	0.09	10	----	----
⁵⁴ Mn	312d	A	6.5	7.4	3.1	3.20	----	256	165
⁵⁸ Co	71d	A	----	0.48	1.7	0.59	----	----	----
⁶⁰ Co	5.24yr	A	1.6	1.4	0.85	0.079	19.4	9.18	2.2
⁶⁵ Zn	243d	A	15	11	9	0.037	----	----	37.8
⁸⁸ Y	107d	A	0.009	1.1	0.031	0.006	----	17.0	----
^{110m} Ag	260d	A	0.42	0.22	0.023	0.043	3.3	----	1.53
¹²⁴ Sb	60.2d	A	0.23	1.7	0.20	0.014	----	22.1	----
¹³⁴ Cs	2.1yr	A	0.91	0.37	0.31	----	6.0	----	----
²⁰⁷ Bi	30yr	A	----	----	0.043	0.006	6.5	----	----
⁹⁵ Zr-Nb	65d	F	300	450	1000	360	2380	4730	7940
¹⁰³ Ru	40d	F	180	330	250	17.6	4130	8520	4350
¹⁰⁶ Ru	365d	F	50	99	62	14	510	1990	1500
¹²⁵ Sb	2.7yr	F	35	43	5.4	8.2	311	960	69.8
¹³⁷ Cs	30yr	F	63	120	55	22	567	2820	1640
¹⁴⁰ Ba	12.8d	F	220	21	540	N.D.	12,800	2030	13,200
¹⁴¹ Ce	32.5d	F	----	280	470	31	----	----	----
¹⁴⁴ Ce	285d	F	150	160	270	94	3360	1230	5360

1. Air is sampled bimonthly. Table values represent the average during the rain and/or flora collection period.

2. C is cosmic ray produced radionuclide
 N is natural radionuclide
 A is activation produced radionuclide
 F is fission produced radionuclide

Radionuclide Concentration Factors in Air, Rainwater, and Flora
March 1967 - Upper Hoh River

SAMPLE			C.F. RAINWATER	C.F. LICHEN ⁽¹⁾ RAINWATER	C.F. MOSS ⁽³⁾ RAINWATER	C.F. MOSS ⁽²⁾ RAINWATER	C.F. MOSS ⁽⁴⁾ RAINWATER	C.F. GRASS ⁽⁵⁾ RAINWATER	C.F. GRASS ⁽⁶⁾ RAINWATER	C.F. FERN ⁽⁷⁾ RAINWATER	C.F. WATERCRESS RAINWATER	C.F. SPRUCE ⁽⁸⁾ RAINWATER
			3/21	3/21	3/21	3/21	3/21	3/21	3/21	3/21	3/21	3/21
DATE	CANOPY		OPEN	MIXED	CLOSED	CLOSED	CLOSED	OPEN	OPEN	CLOSED	CLOSED	OPEN
UNITS	t _{1/2}	SOURCE ⁽⁹⁾	10 ⁵	10 ³	10 ³	10 ³	10 ³	10 ³	10 ³	10 ³	10 ³	10 ³
⁷ Be	53d	C	42	0.33	0.013	0.25	0.56	1.24	0.44	0.25	0.031	0.54
²² Na	2.6yr	C	61	----	0.081	0.22	0.75	0.29	0.27	0.12	0.16	0.046
⁴⁰ K	1X10 ⁹ yr	N	----	11	6.5	4.9	----	----	----	----	----	----
⁵⁴ Mn	312d	A	35	2.9	1.8	5.8	11.0	2.5	4.8	1.7	0.21	3.3
⁶⁰ Co	5.24yr	A	6.8	16	2.5	12	15	2.1	3.5	1.5	0.57	1.7
⁸⁸ Y	107d	A	15	1.1	0.15	1.1	2.2	1.8	2.2	0.74	0.082	1.1
⁹⁵ Zr-Nb	65d	F	11	1.8	0.30	2.4	4.0	2.8	3.7	1.3	0.16	2.1
¹⁰³ Ru	40d	F	26	0.19	0.043	0.26	0.59	0.63	0.46	0.28	0.04	0.41
¹⁰⁶ Ru	365d	F	20	2.8	0.48	2.6	3.2	0.57	0.68	0.41	0.11	0.25
¹²⁴ Sb	60.7d	A	13	----	----	1.2	----	1.7	2.0	----	----	1.1
¹²⁵ Sb	2.7yr	F	22	3.2	1.4	5.0	4.9	----	1.2	0.55	0.10	0.31
¹³⁷ Cs	30yr	F	23	14	3.8	7.2	12	1.7	3.1	1.30	0.10	1.4
¹⁴⁰ Ba	12.8d	F	98	2.7	0.018	1.8	0.82	2.4	0.87	0.51	0.009	0.64
¹⁴⁴ Ce	285d	F	7.8	7.1	2.0	9.4	14.0	3.6	5.4	3.4	0.50	2.9

1. *Lobaria Oregana* - Epiphyte
2. *Selaginella Serpens* - Epiphyte
3. *Sphagnum Mix* - Log Top
4. *Sphagnum Mix* - Ground Cover
5. Saw Grass
6. Meadow Grass - Mixture

7. *Polystichum Munitum* - Sword Fern
8. Sitka-86 Growth
9. Source of Radionuclide
 1. Cosmic Ray = C
 2. Naturally Occurring = N
 3. Neutron Activated = A
 4. Fission Product = F

Subcritical Multiplication of ²⁵²Cf
Neutrons and Its Application

N. A. Wogman and R. W. Perkins

Subcritical multiplication of ²⁵²Cf neutrons has been successfully demonstrated to show that it can be accomplished in a straightforward and safe manner. This accomplishment provides solutions to two problems which presently hinder some of the most useful potential applications of ²⁵²Cf. First, it permits high neutron fluxes to be achieved without resorting to extremely large and expensive ²⁵²Cf sources; and second,

it permits a 10- to 100-fold reduction in radiation levels for a given shield during the transportation of portable sources. Neutron flux amplifications of some 40-fold have been readily achieved and with proper source sizes will provide fluxes which are adequate for many applications of neutron activation, neutron radiography, and other uses normally considered the domain of reactors. A subcritical demonstration facility consisting of PuO₂-polystyrene having a k_{eff} of 0.978, was assembled. At distances of 15 cm and greater where most neutron exposures would be made

(areas of fairly uniform flux), the flux was enhanced by more than 35-fold over that of an unmultiplied ^{252}Cf source. Many neutron activation analysis techniques use a thermal neutron flux of $10^{13}\text{n}\cdot\text{cm}^{-2}\cdot\text{sec}^{-1}$ on a 100 mg sample. Comparable induced radioactivity could be produced with $10^{10}\text{n}\cdot\text{cm}^{-2}\cdot\text{sec}^{-1}$ and 100 g samples and this is entirely feasible for many sample types. Utilizing a subcritical multiplier design with a $k_{\text{eff}} = 0.975$ and optimum flux trap techniques, a 100 mg ^{252}Cf source would provide 2 to 3×10^{10} thermal neutrons $\cdot\text{cm}^{-2}\cdot\text{sec}^{-1}$.

To demonstrate the feasibility and application of subcritical multiplication, a 200 μg ^{252}Cf source was employed in the above PuO_2 - polystyrene assembly. Samples of molybdenum ore concentrate, coal and several biological materials were irradiated. A one hour irradi-

ation of a 66 g specimen of molybdenum concentrate and subsequent gamma-ray analysis of the resultant radionuclides with a 4.2% Ge(Li) diode, allowed 16 elements to be analyzed. Some elements such as rhenium were present only at the ppm levels. A much larger primary source of neutrons (10 to 100 mg of ^{252}Cf) would be adequate for analysis of trace constituents in many large biological specimens. The high flux would also be necessary to analyze trace constituents such as mercury, selenium, antimony and others at the ppm to ppb level in coal.

Additional important applications of neutron multiplication are in producing short-lived isotopes for industry, research and medicine. Neutron multipliers could also provide the necessary thermal flux for a variety of industrial process control applications where conventional methods are not feasible.

Neg 715498-8

Gamma-ray Spectra of Neutron Activated Stibnite

Neg 715498-3

Subcritical Multiplying Demonstration Assembly

TRACER STUDIES IN THE METROMEX EXPERIMENT

The central objective of the METROMEX project is to study convective storms in the St. Louis metropolitan area. During August of 1971 Battelle-Northwest personnel released ruthenium, selenium and gold tracers into the updrafts of convective storms from a ground-based aerosol generator. Samples of rain from these storms, collected using a network of over 100 samplers, are being analyzed for these tracers by neutron activation to determine aerosol scavenging efficiencies and aerosol and hydrometeor trajectories. Large volume rain samples were analyzed for the short-lived cosmogenic radionuclides ^{24}Na , ^{38}Cl , and ^{39}Cl in order to determine aerosol scavenging efficiencies and the elapsed times between scavenging and the arrival of the precipitation at ground level. Air filter samples collected from ground level to 45,000 feet both upwind and downwind of St. Louis are being analyzed to determine the amounts of contaminating materials added to the atmosphere by the St. Louis metropolitan area.

Preliminary Report on Experiments Conducted by Battelle-Northwest During METROMEX

J. A. Young, T. M. Tanner,
C. W. Thomas and N. A. Wogman

Field experiments in the METROMEX project were conducted by investigators from several organizations during the summer of 1971 in a 2000 square mile area centered around St. Louis, Missouri. It is planned to continue the experiments for the next four summer seasons. The central objective of this project is to study convective storms, including their dynamics and microphysics. Special attention is being given to determining the effects of the heat and contaminating materials added to the atmosphere from the St. Louis area on precipitation processes and aerosol scavenging in convective

storms. The participation of the Radiological Chemistry Section of Battelle-Northwest in the METROMEX experiment has three main objectives. The first is to determine by means of neutron activation and X-ray fluorescence analysis the contribution of the St. Louis metropolitan area to the trace element composition of air and rain. A second objective is to add six inert tracers to convective storms in order to determine scavenging efficiencies and aerosol particle and hydrometeor trajectories by means of neutron activation analysis of precipitation samples. The third objective is to determine aerosol scavenging efficiencies and times between scavenging and arrival of the precipitation at ground level by measurements of cosmogenic radionuclide concentrations in precipitation.

Battelle personnel conducted field experiments for the METROMEX project from August 1 through August 20 during 1971. RB-57 aircraft were used to collect air filter samples at 3, 6, 9, 12 and 15 thousand feet both upwind and downwind of St. Louis to determine the contribution of the St. Louis metropolitan area to the atmospheric aerosol. Samples were collected two days a week on each of the first three weeks of August. Air samples were also collected for 24 hour sampling periods using hurricane air samplers at Scott Air Force Base and at a farm southwest of St. Louis (see Figure). Normally the farm is upwind of St. Louis and Scott AFB is downwind. Anderson and Lundgren impactors were used to separate airborne particulates into different particle size ranges. These samples were brought back to the Richland laboratory for trace element analysis by neutron activation and X-ray fluorescence analysis.

Tracers were released into the atmosphere from ground level by means of an aerosol generator which produces submicron sized particles of a tracer material by burning acetone solutions of the tracer. The tracers were released for 30 or 40 minutes immediately preceding the arrival of a convective cell at the generator site, in an attempt to introduce the tracers into the updraft of the cell. Ruthenium, gold and selenium were released

sequentially during the 40 minutes prior to the arrival of a cell soon after midnight on August 11. Ruthenium was released prior to the arrival of a convective cell on the afternoon of August 14, and gold was released during the evening of August 14. Following each tracer release, samples of rain were collected using a network of over 100 rain samplers in the St. Louis area. The samples were frozen and returned to the Richland laboratory where they are currently being analyzed in an attempt to determine the trajectories and precipitation scavenging efficiencies of the tracer materials.

Large volume rain samples were also collected for short-lived cosmogenic radionuclide analysis by means of a 1590 ft² tarp located at Scott AFB. Unfortunately, rain fell at Scott AFB during the experimental period only on August 14. The activities of the short-lived cosmogenic radionuclides in the rain were similar to those measured at Quillayute, Washington, suggesting that the cosmogenic radionuclides present in the rain came primarily from low altitudes and did not enter the convective cell at mid-tropospheric levels. Cosmogenic radionuclide activities are two orders of magnitude higher at mid-tropospheric levels than at ground level.

Neg 720282-3

METROMEX Rain Collection Network

Chemical Characterization of Urban Aerosol

L. A. Rancitelli and T. M. Tanner

The atmospheric aerosol is derived from a variety of natural and man-made sources, each with a particular chemical composition. The major natural sources include the sea salts from the oceans and terrestrial dust derived from the contents. The anthropogenic sources of the atmospheric aerosol are too numerous to tabulate, but can in general be ascribed to major population centers. It has become evident through our studies of the chemical composition of the high altitude aerosol and rain water collected on the west coast of the State of Washington, that the northern hemisphere concentrations of a number of elements have been increased by orders of magnitude as a result of man's activities. Thus, any model describing the mechanisms responsible for the distribution, transport and deposition of aerosol must consider the material emanating from urban centers. In an effort to characterize the chemical composition of urban aerosols, a highly sensitive and precise technique of instrumental neutron activation analysis was developed that is capable of simultaneously measuring over 30 elements on air filters used to sample the urban aerosol at ground level.

The technique involves a one-minute irradiation of the filter samples together with the appropriate standards at a neutron flux of 1.3×10^{13} n/cm² sec. The sample and standards are allowed a 10-minute decay interval to reduce interference from ²⁸Al and then counted for 5 minutes on a Ge(Li) spectrometer. This permits the simultaneous measurement of Al, Br, Cl, Mg, Mn, Na, Ti and V. The samples and standards are then re-irradiated to an integral neutron flux of 10^{17} n/cm², allowed to decay for 5 and 20 day periods, and counted on a Ge(Li) spectrometer for 30 and 100 minutes, respectively. The 30-minute count permits the measurement of Na, K, La, As, Br, Sm, U and Au, while the 100-minute count provides the concentration of Fe, Co, Sc, Eu, Th, Cr, Sb, Rb, Ag, Se, Cs, Hf, Tb, Te and Hg.

The filter samples which are placed in clean polyethylene capsules for the irradiation are 30 cm² sections taken from a total area of 400 cm² of a Whitman 41 filter paper. The volume of air passed through these filters is usually in the range of 1000-1500 m³. While the results are preliminary, several important observations concerning the origin of elements in the urban aerosol can be inferred from the ratio of trace element concentrations. Our high altitude trace element studies indicate

that the Sc content is a valid baseline indicator of natural earth's crust material. When the trace element concentrations are normalized to the scandium content of a filter and compared with similar ratios in major earth's crust materials such as granite or diabase, it is evident that

the As, Sb, Zn, Hg and Se levels have been increased by several orders of magnitude, while Fe, Mn and Cr show enrichments of an order of magnitude in urban centers. Elements such as La, Eu, Hg, Fe, Al and Co appear to exist at levels compatible with an earth's crust origin.

RADIONUCLIDE DECLINE IN COLUMBIA RIVER WATER

The radionuclide depletion processes in the Columbia River are being characterized following the closure of the Hanford reactors. Residual radioactivity is associated mainly with sedimentary deposits behind McNary Dam. The most abundant radionuclides are ^{55}Fe , ^{65}Zn , ^{46}Sc , ^{155}Eu , ^{60}Co , ^{152}Eu and ^{154}Eu . These radionuclides are bound to the sediments and enter the river by sediment resuspension and dissolution into soluble forms. The main mechanism for depletion of radioactivity from the river, other than radioactive decay, is by scouring of surface sediments during high river flow, and their subsequent transport into the Pacific Ocean. It appears that eventually the radioactive sediment deposits behind McNary Dam will become covered by new, uncontaminated silt deposits. The high fraction of radionuclides in the particulate form in Columbia River water changes their availability for biological uptake.

Transport and Depletion of Radionuclides in the Columbia River

D. E. Robertson, W. B. Silker
and J. C. Langford

With the closure of the KE plutonium production reactor in January, 1971 essentially no additional radioactivity has been directly discharged to the Columbia River in reactor effluent water. However, some residual radioactivity has remained in the river, being associated mainly with the sediments and the resuspended load. The Columbia River is, therefore, an extremely valuable natural laboratory for studying the release of radionuclides from river sediments, and for evaluating the natural cleaning processes of river systems following intentional or accidental releases of radioactivity. In addition, these radionuclides serve as excellent tracers for characterizing the

rates and mechanisms of sediment transport in the Columbia River system.

In April, 1971 sampling programs were initiated to study the distribution and behavior of radionuclides in Columbia River sediments, suspended matter, and water between Hanford and Bonneville Dams.

Sediment cores were taken behind McNary and The Dalles Dams using a 6 in. diameter gravity corer. No large sediment deposits could be located behind John Day and Bonneville Dams. The cores were frozen immediately after collection and then sliced into 2.5 cm thick slabs, which were trimmed, oven-dried, homogenized and placed in 4 in. diameter by 1/2 in. thick plastic counting dishes. The sediments were counted on a multidimensional gamma-ray spectrometer and an anticoincidence shielded Ge(Li) diode detector.

A typical gamma-ray spectrum of McNary Dam surface sediments, as counted on an anticoincidence shielded Ge(Li) detector, is shown in the accompanying figure. Ten radionuclides of Hanford origin are present in the sediments including ^{46}Sc , ^{54}Mn , ^{60}Co , ^{65}Zn , ^{125}Sb , ^{137}Cs , ^{144}Ce , ^{152}Eu , ^{154}Eu and ^{155}Eu . Iron-55 was measured in some sediments after radiochemical separations and was found to be the most abundant radionuclide. The typical radionuclide composition of surface sediments behind McNary Dam is shown in an accompanying illustration. Although ^{55}Fe is the most abundant radionuclide in the sediments, it emits only 5.9 keV X-rays and Auger electrons during its disintegration, and from external dose considerations is of minor importance. Analyses of selected core samples for plutonium radionuclides are in progress.

The depth distribution of seven radionuclides of Hanford origin in the sediments behind McNary Dam is of particular interest. The concentration of the shorter half-life radionuclides ^{46}Sc , ^{54}Mn and ^{65}Zn decrease rapidly with depth in the core, whereas the long-lived components ^{60}Co , ^{137}Cs , ^{152}Eu , ^{154}Eu and ^{155}Eu can be found at a depth of 50 cm at concentrations comparable to surface values. The peak in radionuclide concentrations at a core depth of about 28 cm corresponds to a maximum release of radioactivity to the river in June, 1967. The sediment deposition rate at this core location behind McNary Dam between June, 1967 and April, 1971 can then

be extrapolated to be about 7 cm/year. Sediment deposition rates calculated from changes in the ratios of $^{46}\text{Sc}/^{65}\text{Zn}$ and $^{65}\text{Zn}/^{60}\text{Co}$ with depth in the sediment core give 7 cm/year and 10 cm/year, respectively.

Because of the very low concentrations of radionuclides in Columbia River water, Battelle Large Volume Water Samplers were used to remove particulate and soluble forms of radionuclides from 500 to 1000 liters of river water. Particulate material was removed on a series of 12 in. diameter fiberglass filters, whose retention efficiencies were shown to be comparable with 0.3 μ membrane filters. The filtered water was then passed through 12 in. diameter by 1 in. thick beds of cation and anion exchange resins to remove dissolved ionic species, and finally through a 12 in. diameter by 1/4 in. thick bed of activated aluminum oxide to remove non-ionic constituents. The flow rate through the filtration-sorption assembly was maintained at 9 liters/minute. The filters, aluminum oxide beds and aliquots of the cation and anion beds were counted directly on high sensitivity multi-dimensional gamma-ray spectrometers to measure the radionuclide concentrations contained on each material.

River water is periodically sampled from the generator cooling supply at McNary, John Day, The Dalles and Bonneville Dams. These sampling locations within the dams were chosen because the generator cooling waters represent vertically mixed water from behind the dams, and are therefore more characteristic of the dynamic flow of the river.

The relative distribution of radionuclides in the Columbia River between particulate matter ($>0.3\mu$), cationic, anionic and non-ionic species indicates that nearly all of the radioactivity in the water is present in the form of resuspended sediments (see table). These radionuclides remain very tightly bound to the sediments, and any leaching of radionuclides from the sediments to form soluble species is very small. It is interesting to note that the ^{106}Ru present in the river as fresh fallout is mainly in a non-filterable form, which is retained on the Al_2O_3 bed. This would indicate that the ^{106}Ru is present in either a nonionic form or a colloidal species.

The transport of ^{65}Zn on resuspended sediments between McNary and Bonneville Dams is shown in an accompanying illustration. Most of the sediments behind McNary Dam are of Snake River origin and are deposited mainly on the Oregon side of the river. Because of this influence of Snake River water and sediments, it was necessary to sample from both the Oregon and Washington sides at McNary Dam in order to differentiate the contribution of Snake River water to the Columbia River. The ^{65}Zn concentrations in the suspended matter collected from both sides of the dam clearly show this influence. The Snake River flow began peaking around the middle of March and increased the scouring of the main deposits of radioactive sediments on the Oregon side of the river behind McNary Dam. The higher radionuclide concentrations in the resuspended sediments on the Oregon side of the river became less

pronounced after the Columbia River flow had peaked during the first week in April. The transport of ^{65}Zn (and other radionuclides) on resuspended sediments between McNary and Bonneville Dams indicates no major systematic redeposition of these suspended sediments in the slack water pools behind the dams downstream from McNary. It appears that once surface sediments have been resuspended during the spring freshet, the major portion of them is flushed completely out of the river and into the ocean. The absence of any major deposits of fine sediments between McNary and Bonneville Dams supports this observation. With the recent construction of John Day Dam some sedimentation must occur in the slack water behind this dam, but the transport rates of resuspended radioactive sediments indicates this is not a major sedimentation basin.

As indicated in the accompanying illustration, there is a direct relationship between river flow rate and transport rates of radionuclides past Bonneville Dam. The rapid increase in radionuclide transport correlated with the spring freshet, and then decreased during the period in which river flow remained at a maximum. Apparently the spring freshet immediately scours off a layer of the finest surface sediments and then resuspends the underlying sediments at a slower rate. The rapid decrease in the radionuclide transport rate closely followed the decline in river flow rate, and then leveled off after August when the low river flow stabilized. The total curies of radioactivity of Hanford origin (excluding

^{55}Fe) transported past Bonneville Dam and presumably into the Pacific Ocean between February and December, 1971 is estimated to be about 600 curies. This may be compared with annual discharges on the order of 350,000 curies/year during the early 1960's when eight production reactors were operating.

The main mechanism for depletion of radionuclides (other than radioactive decay) from the Columbia River is by the resuspension of surface sediments during the spring freshet and high water runoff, and the sub-

sequent transport of the suspended sediments to the ocean. Since the net deposition rate of new, nonradioactive sediments (mainly of Snake River origin) behind the dams is apparently greater than the scouring losses of old sediments by high runoff, it is probable that the radioactive deposits will eventually become buried by fresh layers of uncontaminated silt. The extent of this burial and the resulting decrease in river concentrations will be defined as this study progresses.

PERCENTAGES INDICATE THE FRACTION OF THE TOTAL ARTIFICIAL RADIONUCLIDE CONTENT OF THE SEDIMENTS AND THE NUMBERS IN PARENTHESES ARE THE TYPICAL CONCENTRATIONS IN D/M/g OF DRY SEDIMENT

Neg 720292-1

Typical Composition of Photon Emitting Radionuclides in Surface Sediments Behind McNary Dam, April, 1971

Iron-55 Concentrations in
Columbia River Sediments, 1971
Units of D/M/g

Sediment Depth, cm	McNary Dam - Oregon Side
Surface	1120 ± 30
30	172 ± 6
43	178 ± 7
48	366 ± 9
	McNary Dam - Washington Side
8	1250 ± 30
10	701 ± 18
13	460 ± 13
15	613 ± 18
18	385 ± 13
20	291 ± 11
	The Dalles Dam
5	87 ± 3
8	98 ± 4
10	70 ± 3
15	20 ± 2

Neg 713401-1

Columbia River Surface Sediment--McNary Dam

Neg 715761-2

Depth Distribution of Hanford Origin Radionuclides in
Columbia River Sediments Behind McNary Dam
April, 1971

Typical Distribution of Physico-Chemical Forms of Radionuclides
in the Columbia River - McNary Dam, July 13, 1971

Percentage Distribution on Filters, Resins and Al_2O_3

<u>Unit 2 - Oregon Side</u>	<u>^{46}Sc</u>	<u>^{60}Co</u>	<u>^{65}Zn</u>	<u>^{106}Ru</u>	<u>^{152}Eu</u>	<u>^{228}Th</u>	<u>^{226}Ra</u>
Particulate ($>0.3\mu$)	100.00	98.1	99.0	0	98.4	70.0	99.9
Cationic	0	1.3	0.6	0	1.6	2.5	0.1
Anionic	0	<0.3	<0.1	0	0	0	0
Nonionic	0	0.6	0.5	100.00	0	27.5	0
<u>Unit 14 - Washington Side</u>							
Particulate ($>0.3\mu$)	99.7	98.6	99.3	39.4	97.3	100.0	100.0
Cationic	0	9.7	0.4	0	1.1	0	0
Anionic	0	0	0	0	0	0	0
Nonionic	0.3	0.7	0.4	60.6	1.6	0	0

Neg 715761-3

Transport of ^{65}Zn in the Columbia River Between McNary Dam
and Bonneville Dam

Neg 715761-4

Transport of Radionuclides* Past Bonneville Dam
April Through November, 1971

RADIOLOGICAL PHYSICSDOSIMETRY 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. B. Shipler,* J. E. Ballou,**
D. McConnon*** and I. C. Nelson

Efforts continued toward development of a method for diagnosing the retention of promethium in the bodies of workers following accidental inhalation of $^{147}\text{Pm}_2\text{O}_3$. Ultimately a quantitative diagnostic method is expected to be obtained as a result of human testing using promethium-143 in place of promethium-147 in an oxide aerosol. Promethium-143 facilitates in vivo detection with low doses to participating volunteers. However, the mass of promethium-143 based on detection capability and acceptable dose is inadequate for the formation of realistic aerosols. Since there is no stable isotope of promethium, samarium is under investigation as a carrier for promethium.

Experiments involving intravenously injected promethium-147 and samarium-145 chlorides in citrate buffer solution in rats disclosed that no gross differences occurred in the metabolism of the two radionuclides in that species.⁽¹⁾

During the year efforts centered on assessing the feasibility of using samarium as a carrier for promethium-143 in inhalation studies using the rat. Thirty-six rats were exposed (nose only) to an aerosol consisting of particles of $^{143}\text{Pm}_2\text{O}_3$ - $^{145}\text{Sm}_2\text{O}_3$ and Sm_2O_3 having an aerodynamic median activity diameter of 3.4 μm and a geometric standard deviation of 1.6. One group was sacrificed immediately and the lungs were analyzed to determine the initial lung burden.

Periodic collections of urine and feces were made for each rat. Groups of rats were sacrificed at 14 and 30 days following exposure and lungs, liver, kidneys, head, muscle, pelt and skeleton were sampled at necropsy. The samples were analyzed using coincidence gamma scintillation counting. All samples for which analysis

* Present address: Oregon Technical Institute, Klamath Falls, OR.

** Inhalation Toxicology Section, Biology Dept., Battelle-Northwest

*** Present Address: Rural Cooperative Power Assoc., Elk River, MN

was intended have been analyzed. Numerical analysis of the data continues. Preliminary results indicate that the ratio of samarium to promethium in tissue samples agrees favorably with the ratio of the nuclides in the aerosol and suggests that samarium will be usable as a carrier for promethium-143 in inhalation studies.

Plutonium

P. J. Dionne* and I. C. Nelson

A computer model predicting the excretion, translocation and retention of plutonium in the human body following intravenous injection of plutonium nitrate was developed through comparison of such data obtained by Langham, et al.⁽²⁾ for plutonium citrate in humans, Stover, et al.⁽³⁾ for plutonium citrate in beagle dogs and Bair, et al.⁽⁴⁾ for plutonium nitrate in beagle dogs. Recognizing the possible limitations of the model due to differences in pH, chemical composition and amounts of the injected material, prediction factors were estimated relating results of the human and dog models for injected plutonium citrate. These prediction factors along with the dog model for plutonium injected in nitrate form were used to develop predictions of initial whole body, bone and liver burdens given the daily amount of plutonium excreted in urine for injection of plutonium nitrate in humans. Present results, which are subject to revision as the

model is refined, suggest that the total body burden of plutonium following intravenous injection of plutonium nitrate can be estimated from

$$B_{TB} = 2.4 \times 10^3 U \exp (0.693t/1.3) \quad 1 \leq t \leq 8$$

where B_{TB} is the total body burden in the same units as the daily urine excretion, U , and t is time in days. Similarly, the bone burdens can be estimated from

$$B_{Bone} = 1.9 \times 10^2 U \exp (0.693t/1.3) \quad 1 \leq t \leq 8.$$

For comparison, the data presented by Langham, et al.,⁽²⁾ for injection of plutonium citrate suggests a total burden of $B = 4.3 \times 10^2 U t^{0.77}$ $0 \leq t \leq 136$.

Further study and manipulation of the model is expected to yield estimates of the most probable organ burdens over longer time intervals.

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* Systems Engineering Department, Battelle-Northwest.

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 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.

Chemical Reactivities of Radicals In Aqueous Solution

D. R. Kalkwarf, H. W. Fenrick*
and R. N. Diebel

A systematic study of relations between the structures and reactivities of radiation-induced biochemical radicals in aqueous solution was continued. Radicals were formed in irradiated crystals of biochemical compounds or irradiated samples of their solutions in ice since in these forms their concentrations and general structural features could be readily determined by electron spin resonance. Bleaching of erio-glaucline at pH 6 and of p-nitrosodimethylaniline at pH 9 were again used to detect the presence of oxidizing radicals, while one-electron reduction of flavin mononucleotide was used to detect reducing species.

The most numerous class of radicals found in irradiated biochemical crystals is the carbon radical, i.e., a radical in which the unpaired electron is predominantly associated with a particular carbon atom in the molecule. Radicals containing a carboxylate group adjacent to this carbon atom were found to be particularly potent oxidizing agents. Examples include $^{\ominus}\text{OOC}-\dot{\text{C}}\text{H}-\text{COO}^{\ominus}$ derived from malonic acid, $^{\ominus}\text{OOC}-\text{CH}_2-\dot{\text{C}}\text{H}-\text{COO}^{\ominus}$ derived from succinic acid, and $\text{CH}_2-\text{COO}^{\ominus}$ derived from glycine. When positively-charged or other electron-withdrawing groups were present in the radical, oxidizing ability was greatly decreased. This was illustrated by $\text{OOC}-\dot{\text{C}}(\text{OH})(\text{COOH})-\text{CH}-\text{COO}^{\ominus}$ derived from citric acid, $\text{H}_3\text{N}^{\oplus}-\text{CH}_2-\dot{\text{C}}\text{H}-\text{COO}^{\ominus}$ derived from β -alanine and by most of the $-\text{C}(\text{OH})-$ radicals derived from natural sugars.

The possibility of structural rearrangement in biochemical radicals during hydration was examined by

* Visiting Professor Appointment,
Summer 1972, from Wisconsin State
University.

comparing the ESR spectra and chemical reactivities of radicals formed in both dry crystals and in 1M ice solutions irradiated at -196°C . For many of the samples investigated, e.g., glycine, α -alanine, lactose, valine, ESR spectra of the radicals formed in ices were either very similar to those seen in irradiated crystals of the compound at room temperature or became similar as the ice was warmed. Exceptions were found, however, e.g., maltose and fructose. Whether formed in irradiated crystals or ices, radicals derived from α -alanine, galactose and maltose were found to be oxidizing agents while the radicals derived from β -alanine were found unreactive toward the diagnostic reagents. Until more extensive comparisons can be made, it was concluded that hydration generally does not alter the chemical structures of biochemical radicals and that their reactivities reported here and in past reports should be applicable in irradiated living tissue.

Radical Reactions of Biochemical Membranes

D. R. Kalkwarf

Research was begun to test whether free-radical attack at biochemical membranes could be a particularly significant means for amplifying radiation damage in living systems. A model membrane system was developed⁽¹⁾ which can be made extremely sensitive to changes in chemical structure and should be a good indicator of free-radical or other types of radiation damage. This membrane consists of a

protein-activated lipid bilayer separating two aqueous compartments, and the electrokinetic phenomena occurring at the interface can be made to closely resemble excitable cell membranes.

In preparation for these experiments, ESR spectrometry and diagnostic reagents were used to detect the presence and reactivities of any radicals formed in gamma-irradiated crystals of the membrane-forming lipids and related compounds. Irradiated glycerides, e.g., tripalmitin, the various dipalmitins and tristearin, were found to form $\text{R}_1\text{-CH}_2\text{-}\dot{\text{C}}\text{H-COOR}_2$ radicals. In contrast, irradiated phospholipids such as β,γ -dipalmitoyl- α -lecithin and stearyl-sphingomyelin gave no ESR signals. Steroids such as progesterone, cholesterol and cholesterol acetate gave complex spectra which have yet to be interpreted.

Radical-induced oxidation of diphenylpicrylhydrazyl in chloroform and polymerization of vitamin E in benzene were used to evaluate the reactivities of these radicals. All of the radicals detected by ESR spectrometry were found to bleach diphenylpicrylhydrazyl. Only radicals derived from dipalmitin and tripalmitin have been tested as yet with vitamin E, but both of these were found to dimerize the reagent as detected by the increased optical absorption at 305 nm. These experiments show that radicals derived from naturally occurring lipids can extend the effects of radiation damage and that their behavior in lipid membranes could be of great significance in the development of this damage.

Flash Photolysis of Nucleic Acid Components

W. D. Felix

The reaction kinetics of the radiolysis products of water, i.e., the hydroxyl radical ($\dot{\text{O}}\text{H}$) and the hydrated electron ($\dot{\text{e}}_{\text{aq}}$) have been characterized for many substances of biological interest. In most cases, the products of these reactions are, themselves, highly reactive free radicals. For example, in the biological environment, it would be expected that the products of hydroxyl radical reaction with the chloride or carbonate ions of the body fluids will result in the formation of the free radicals $\text{Cl}\dot{1}_2$ and $\text{CO}\dot{3}$. These radical ions are expected to be strong oxidizing agents and should be expected to significantly contribute to the general disruption of cellular processes as they interact with cellular components.

In this study the reactions of the $\text{CO}\dot{3}$ radical ion are being investigated in order to determine its relative biological damage potential. In preparation for this study, the flash lamp assembly developed in this laboratory has been improved. Because of the complexity of the mechanisms involved, it has also been necessary to develop an effective data handling system. Flash photolysis data are collected directly with a digital memory oscilloscope and transferred to paper tape for analysis, or photographs of oscilloscope traces are analyzed with a semi-automatic reader for conversion of information to punched paper tape. Edit programs then convert the punched paper tape into a computer compatible format.

Simultaneously with this development work, programs were also written to synthesize ESR spectra whereby direct comparison with experimental data may be made for use in the identification and characterization of the radical products.

Reaction of the $\text{CO}\dot{3}$ radical as formed during flash photolysis is indicated by a change in the absorption of light at the 600 nm absorbance peak. Rate constants determined for the decay of $\text{CO}\dot{3}$ in the presence of uracil and cytosine (at $2 \times 10^{-4}\text{M}$ concentration) indicate that $\text{CO}\dot{3}$ is only slightly less active than $\text{Cl}\dot{1}_2$ in these two pyrimidine systems. The rate constants, $0.9 \times 10^7\text{M}^{-1}\text{sec}^{-1}$ and $2 \times 10^7\text{M}^{-1}\text{sec}^{-1}$ for reaction with uracil and cytosine respectively, are approximately one fourth of the magnitude of the constants for the respective reactions with $\text{Cl}\dot{1}_2$.⁽²⁾ These initial results indicate that the reactions of the secondary radicals, such as $\text{CO}\dot{3}$, can be significant in consideration of the overall radiation damage picture. However, three factors mitigate against the inclusion of $\text{CO}\dot{3}$ radical reactions in most systems: 1), the rate constants for reaction of $\dot{\text{O}}\text{H}$ with Cl^- is $1.5 \times 10^{10}\text{M}^{-1}\text{sec}^{-1}$ (2) compared with 1.1×10^6 to 3.7×10^8 (pH dependent) for $\text{CO}\dot{3}$; 2), the rate constants of reaction of $\text{Cl}\dot{1}_2$ with the pyrimidines are four times larger than those of $\text{CO}\dot{3}$; and 3), the concentration of Cl^- in the usual cellular fluids is much higher than that of $\text{CO}\dot{3}$. Thus, in most biological systems, reaction of $\text{Cl}\dot{1}_2$ with cytosine and uracil will be significantly more important than the reaction of $\text{CO}\dot{3}$.

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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. Advanced instrumentation concepts, detectors and circuitry are also developed for use in the fields of health physics and environmental studies.

Absolute Measurement of Total Body Calcium by In Vivo Neutron Activation Analysis

H. E. Palmer

with

W. B. Nelp, J. D. Denney, R. Murano,
T. G. Rudd, University of Washington
Medical School, Seattle, Washington.

A method for measuring serial changes in total body calcium in man has been in use for approximately two years. The uniformity of activation, precision, and reproducibility of the measurement are such that a 2% change in calcium content is detectable.^(1,2) With variations among people in body geometry and the amount of tissue moderator surrounding the skeleton, absolute determinations of total body calcium were possible with an accuracy of ± 8 to 10%. In order to improve the absolute accuracy of this method, five cadavers of various body sizes were accurately irradiated and counted several times. The complete skeletons were then carefully ashed by slow heating in a large muffle furnace after which the calcium content of the ash

was accurately determined by an atomic absorption method. The milligrams of calcium per gram of bone ash were remarkably constant from skeleton to skeleton, averaging $366 \text{ mg} \pm 1.9 \text{ mg S.D.}$

As expected, it was found that for any given height, skeletal activation would be somewhat less in the heavier subject as compared to a leaner one. The skeletal activity of the ^{49}Ca was correlated with an expression of "body form" relating the weight per unit height. This expression is weight in kilograms divided by height in meters, i.e., kg/m. There is a linear relationship over the range of cadaver body sizes studied. By using this graph, one can enter the body form, kg/m of any subject, determine the predicted specific activity based on the chemically analyzed series of cadaver standards and then calculate the body calcium. Statistical analysis of the data permits estimation of the accuracy for measuring body calcium in grams in any given individual. By incorporating the known counting errors and the previously determined precision of a single activation,

with the data in the coefficient of variability for estimating body calcium from a single measurement is approximately 5.2%.

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Relation of Specific Activity to Body Form

In Vivo Production of ^{37}Ar and ^{41}Ar From Fast Neutron Irradiation of Calcium

H. E. Palmer

^{37}Ar and ^{41}Ar can be produced by an n, α reaction of fast neutrons with ^{40}Ca and ^{44}Ca respectively. In ani-

mals or humans the ^{37}Ar is produced only from ^{40}Ca and there are no interfering reactions but ^{41}Ar is produced also from ^{41}K in the body by an n,p reaction. ^{37}Ar decays by electron capture, has a 34.4 day half-life, and emits only 2.6 keV X rays and Auger electrons which makes measurement external to the body very difficult. ^{41}Ar undergoes beta decay with a 1.83 hour half-life and emits a 1.29 MeV gamma ray. External measurement can be made only with Ge(Li) detectors which are not very efficient because of their small size. NaI(Tl) detectors cannot be used because the photon radiation from ^{24}Na , ^{42}K , ^{38}Cl etc., completely masks the 1.29 MeV peak from ^{41}Ar .

Studies in rats and dogs have shown that the ^{37}Ar and ^{41}Ar can be measured after they have been separated and collected from the expired air of these animals following irradiation with 14 MeV neutrons. For ^{41}Ar counting the expired air is absorbed onto activated charcoal in a liquid nitrogen trap and the counting done on a large NaI(Tl) detector with the ^{41}Ar still in the trap. For ^{37}Ar counting, diver's gas consisting of 80% helium and 20% oxygen is breathed by the animal. The exhaled atmosphere is passed through hot copper to remove oxygen, through CaSO_4 and P_2O_5 to remove water vapor, and then through Ascarite to remove CO_2 . The remaining radioactive argon and helium are passed through liquid nitrogen cooled activated charcoal which absorbs the argon in a purified form. The argon is then transferred internally into a proportional counter

where both the Auger electrons and X rays from the ^{37}Ar are counted with almost 100% efficiency. The background of the proportional counter under the 2.6 keV photopeak is less than 1 count per minute which allows very sensitive determination of the ^{37}Ar .

Many more studies must be done to determine if expired ^{37}Ar can be used as a measure of total body calcium. Preliminary studies provide the following results and conclusions.

The use of ^{37}Ar as a measure of body calcium is much preferred over ^{41}Ar because it has less interference from other elements and its overall counting efficiency is more than 100 times better than that for ^{41}Ar .

Therefore a much lower neutron radiation dose can be used.

Experiments involving the simultaneous irradiation of both a live and dead rat indicate that 85% of the ^{37}Ar is exhaled within 30 minutes after irradiation.

Irradiation of $\text{Ca}(\text{NO}_3)_2$ solutions and subsequent counting of the ^{37}Ar produced indicate that a 0.01 rad dose to an adult human would provide a count rate of 675 counts per hour over a background of 6 counts per hour. A 15 hour count would provide about 10,000 net counts over a background of 90 counts. The proportional counter is very stable and could be used to count the sample for several days in which case the dose could be reduced to 0.001 rad and the ^{37}Ar counted for 6 days to provide a net count of 10,000 over a background of 900 counts.

If it is found that essentially all the ^{37}Ar is expired from the body with-

in an hour, it is expected that this technique will provide an accurate measure of total body calcium. The technique would eliminate the need for a whole body counter and its associated errors due to different body geometries and self absorption. The radiation dose required by this technique could be only 1% of that currently used for the determination of total body calcium by in vivo neutron activation analysis.

The Determination of Cadmium In Vivo
by Measurement of Prompt Gamma Rays
Following Neutron Capture

H. E. Palmer, L. W. Brackenbush
and L. G. Faust

Cadmium can constitute a serious health problem. Exposure to cadmium in industrial as well as general environments has caused serious intoxications in human beings. Cadmium tends to accumulate in the body and most of it becomes deposited in the kidneys and liver. Animal experiments show that excretion of cadmium via urine and the gastrointestinal tract is very low and related to body burden only in a minor degree.⁽¹⁾ A method to measure existing levels in living humans is needed so that cadmium toxemia can be recognized, treated, or prevented.

Since cadmium has a relatively large cross section for thermal neutrons (20,000 barns for ^{113}Cd), prompt gamma ray measurement during low level neutron irradiation of the liver or kidneys may provide a method for determining cadmium concentrations in these organs. The major prompt

gamma ray emitted during neutron capture has an energy of 559 keV. Preliminary studies were conducted using a collimated beam of neutrons from a ^{252}Cf source which irradiated a water phantom simulating the human liver surrounded by body tissue. A 50cc volume Ge(Li) detector having a resolution of about 2 keV for the 559 keV photopeak was used to measure the prompt gamma rays to produce a spectrum obtained during a one hour irradiation of the phantom in which the liver contained a solution of 4 grams of cadmium per liter. Most of the interfering Compton continuum is produced from captured gamma rays from hydrogen and lead. The radiation dose to the liver during this measurement was approximately 10 millirem. As little as 20 mg cadmium per liter has been detected in the phantom and this is the lower limit of detection.

The normal content of cadmium in kidney and liver adult humans is about

10 and 5 mg respectively.⁽²⁾ Assuming that the masses of the liver and kidneys are 1700 and 300 grams, respectively, the normal concentrations are about 3 and 33 mg per kilogram of tissue. The method as described above will not detect these levels, but with improvements in shielding and collimation the sensitivity might be extended down to these concentrations. Toxicity effects of cadmium become manifest at kidney concentrations of about 140 mg per kilogram.⁽³⁾ This method in its present state of development could detect such levels.

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Neg 720089-1

Prompt Gamma Ray Spectrum From Liver Phantom Containing
4 g/l Cadmium

Radiation Instrumentation

K. L. Swinth

Emphasis on the detection of plutonium translocated to the tracheobronchial lymph nodes continued with the evaluation of advanced probes and thorough analysis of counting geometries. Although advanced probes have displayed more uniform angular sensitivity, expected increases in counting efficiency have not been obtained. With the larger area diodes employed in these probes, one obtains large areas of the surface with poor counting

efficiency. With a typical 2 cm long detector, only 1.2 cm of the length could be considered effective in counting the plutonium X rays. Because of this lack of uniformity, detection limits are higher than originally projected.

The minimum detectable amount (MDA) for the tracheobronchial nodes in the human phantom is 8 nCi under optimum counting conditions with a 20 minute counting period. This was with a single 1 cm long detector-not with the most recent three element probe which had a minimum detectable amount of 15 nCi under

similar conditions. To obtain the required uniformity of angular response a multielement probe is necessary. The measured angular sensitivity of the three element probe is uniform to $\pm 35\%$ whereas the sensitivity of a single element probe drops to almost zero at some angles. It was found that a shaping time constant of 2 μsec was optimum with our system and that anti-coincidence operation of the detectors in the three element probe did not significantly reduce the background count rate.

The tracheobronchial lymph nodes of dogs are smaller and more uniform than those of humans; however, dogs were used to aid in the evaluation of the probes. Analyzing experimental data obtained from dogs it was found that an average MDA of 30 nCi was obtained for the middle tracheobronchial node and an MDA of 16 nCi was obtained for the right tracheobronchial node when employing a one minute counting period. Counting sensitivity was consistently higher for the right tracheobronchial node which is apparently in a more favorable counting position. Because of the high plutonium concentrations in the lymph nodes of the dogs, self absorption of the X rays was considered; however, it was found by calculation that less than 10% of the X rays would be absorbed.

Calculations were performed and available materials investigated for optimization of a fiber optics coupled scintillator for the detection of 17 keV X rays from plutonium. Limiting the sensitive area of the photocathode of the phototube along with

improvements in the light transmission of fiber optics should result in increased sensitivity and decreased background rate compared to previous prototype systems. MDA's would then be comparable to the avalanche probes; however, the counting would depend primarily on the detection of 60 keV gammas from ^{241}Am . The system is being developed for both wound counting and in vivo counting of lymph nodes.

Wound visualization studies have centered around optimization of an image intensifier system. NaI(Tl), CaWO_4 , and ZnS(Ag) scintillators have been used on the system with the clearest images resulting from the ZnS(Ag). Although the other scintillators are more efficient, internal reflections and light scattering tend to "wash out" the image. Because one must obtain a large number of events to produce a recognizable image, such a system will be limited to high activity contaminations. A matrix of detectors with larger "resolution elements" or a position-sensitive proportional counter that could be placed in contact with the wound would be more appropriate in most cases.

Measurements of X-ray-alpha ratios were compiled for a number of transuranic isotopes and provided to members of the "Intercalibration Committee for Low-Energy Photon Measurements." Reported intensities for X rays are rare and differences of greater than a factor of three are evident for some low-energy gamma rays that have been proposed for in

vivo plutonium counting. Plutonium whole body counter intercalibration sources are being routed among interested laboratories; however, only a

preliminary analysis of the data has been performed. When the routing of the sources is completed the data will be analyzed in detail.

Neg 712954-2

Scans Along Length of the Detectors in the Three Element Probe Showing Loss of Sensitivity Near Center of Probe. This is due to non-uniformities in the response of the avalanche detectors.

Counting Sensitivity for Right and Left Tracheobronchial Lymph Nodes in Beagle Dogs Based on Post-Mortem Analysis of Tissues

Dog	Counting Sensitivity (cpm/ μ Ci)	
	Middle T-B	Right T-B
329	48.6 $\pm$ 4.2	300
248	47.5 $\pm$ 4.5 ^(b)	168 $\pm$ 10 ^(b)
335 ^(a)	66.3 $\pm$ 4.5	74 $\pm$ 11.5
344 ^(a)	85.1 $\pm$ 5.5	129 $\pm$ 20
Unweighted Average	66.7	124 (without 329)

a. Post mortem experiment

b. Counts with probe D-1, but normalized to probe LA-3

Neg 712954-6

Scans made in a Human Thorax Phantom Showing Increase in Count Rate Near Lymph Node Sources with an Optimum Detector Configuration. The inter-tracheobronchial source is at the 2 cm position and the laterotracheal sources are at 5 cm.

MECHANISMS OF ENVIRONMENTAL EXPOSURE

This project involves a continuation of the environmental study to measure the decline of radioactivity in various population groups and to reduce, compute, analyze and report the environmental information that can be extracted from these new studies and those parts of the completed research not adequately reported.

A Study of Seafood Consumption and ^{65}Zn Body Burden

T. H. Essig, G. W. R. Endres and
J. F. Honstead*

One of the radionuclides formed by neutron activation in Hanford production reactors and released to the Columbia River in effluent cooling water prior to February, 1971 was Zn-65. This radionuclide has a relatively long radioactive half-life (245 days) and is an isotope of an element utilized in metabolic processes by most shellfish and other aquatic life forms. Consequently, Zn-65 is found in fish and shellfish in the Columbia River and in the Pacific Ocean near the mouth of the River. The transfer of ^{65}Zn through various food chains to man has been studied at Hanford for a number of years. (1,2,3,4)

Since the half-life of ^{65}Zn is fairly long, it can enter into rather complex transport processes in a variety of food chains and can persist in measurable quantities in the en-

vironment--particularly in organisms that concentrate zinc by metabolic absorption. Shellfish such as oysters, clams and crabs concentrate zinc (and consequently ^{65}Zn) to a high degree and thus represent the dietary items of primary interest in this study.

When ^{65}Zn decays, a 1.11 MeV photon is emitted. The energy of this photon is high enough to be readily detected by a scintillation crystal external to the object being measured, e.g., environmental samples or the human body. Thus, scintillation counter techniques (such as those employed in a mobile whole-body counter), together with knowledge of an individual's dietary habits, can be used to study the environmental behavior of ^{65}Zn , particularly with those individuals consuming large quantities of shellfish from the Columbia River estuary.

In this study, consumption rates of four seafoods (clam, crab, shrimp and oysters) were obtained for 250 residents of Rockaway, Oregon, and Ilwaco, Washington, by personal interviews. These consumption rates were used in conjunction with ^{65}Zn concentrations in the seafoods of interest

* Deceased

to estimate the ^{65}Zn body burden for each individual. Rather sophisticated equations were used to adjust the seafood intake as a function of age for the three years preceding this study. This adjustment was necessary because of the relatively long half-life of ^{65}Zn . The following equation (adapted from reference 5) was used to calculate the ^{65}Zn body burden (q):

$$q = \frac{f_w I T_e}{0.693} [1 - \exp(-0.693t/T)]$$

where

f_w = fractional uptake of ^{65}Zn for each type of seafood,
 I = intake rate of ^{65}Zn , nCi/day,
 T_e = effective half-life = 194 days⁽⁵⁾
 and
 t = period over which ^{65}Zn was ingested, days.

After each subject was interviewed, he was counted in a mobile whole-body counter located in the vicinity of each of the seacoast communities. The ^{65}Zn body burdens measured by this technique were corrected for background and Compton scattering, and were then compared with the body burdens calculated by Equation 1. The results of this comparison for one group of people (seafood company workers are shown in the figure.

The agreement between the measured and calculated ^{65}Zn body burdens for the seafood company workers is considered to be very good. The average measured-to-calculated ratio of body burdens was 1.47, with a range in individual ratios from 0.11 to 3.7. This range of individual ratios is felt to reflect a combination of natural variability of key metabolic parameters and of the ability of an individual to quantitatively estimate his diet.

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RADIOLOGICAL EVALUATION OF POST MORTEM TISSUE SAMPLES

I. C. Nelson, K. R. Heid,* P. A. Fuqua, MD** and T. D. Mahony, MD***

This study is concerned with the collection, radioanalysis and evaluation of tissue samples obtained post-mortem from plant workers and residents in the environs of the Hanford Project. The significance of this effort lies primarily in developing the capability for assessing the overall long-term effectiveness of engineering safeguards which govern the release of radioactive materials to the Hanford environs. The data are similarly important for assessing the overall impact on occupational workers through correlation of exposure conditions and resulting body burdens.

Results of plutonium analyses on tissue samples obtained prior to late 1970 were assembled, analyzed and reported. Collection of lung, liver, bone and tracheobronchial lymph node samples continued throughout the year. In November, 1971 samples of spleen and gonads were added to the list of desired samples. The presence of plutonium in the spleen has been suggested from animal studies following intravenous or intramuscular injection of plutonium compounds. Positive findings of plutonium in the spleens

of workers may establish the deposition of plutonium as resulting from contaminated wounds. The presence or absence of plutonium in gonadal tissue may be of interest from a genetic point of view.

An expansion of the sampling program to include individuals remote from the Hanford complex and to include analysis for certain selected stable nuclides was discussed at a meeting with representatives of the Los Alamos Scientific Laboratory. Tentatively, Pacific Northwest Laboratory will consider expansion of sampling programs to selected localities west of the Continental Divide whereas the Los Alamos group will emphasize sampling from localities east of the Divide.

* Occupational and Environmental Safety Department, Battelle-Northwest.

** Hanford Environmental Health Foundation, Inc.

*** Kadlec Hospital, Richland, WA

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 distribution of energy deposition with temporal distributions. Emphasis has been on understanding time dependent effects and dose rate dependence of the oxygen effect. Experimental work has been in the application of cross section measurements to the problem of energy transport by δ -rays and to a more detailed description of proton stopping powers.

Energy Transport by Secondary Electrons

W. A. Glass

In order to understand the nature of energy deposition in small volumes traversed by protons, it is essential to know the probabilities involved for energy transfer in individual collisions between the proton and the absorber molecules. An analysis of the relative importance of the various processes by which the energy of fast protons is transferred to the absorber shows that about 50 to 60 percent appears as kinetic energy of secondary electrons, 25 to 30 percent is involved in overcoming the binding energy of these electrons, and the remaining 10 to 20 percent appears as primary excitation, recoil energy, etc.⁽¹⁾

It has been observed that distributions of energy deposited in small volumes by protons are greatly affected by energy transport associated with energetic secondary electrons. The moments of these distributions do

not conform to those expected in the absence of energy transport.⁽²⁾ Quotients of the variance by the mean of ionization distributions measured in small tissue-equivalent sites were found to be nearly independent of proton energy.

The general distribution of those electrons ejected in primary collisions can be put in perspective by considering the number of electrons and the kinetic energy associated with them above certain selected secondary electron energies. The table summarizes data for two representative low Z gases bombarded by protons between 0.3 and 1.5 MeV. The mean ejected electron energy lies between 30 and 60 eV and this value is only slightly dependent on whether the target is hydrogen or nitrogen. Nearly a constant 75 percent of the kinetic energy is associated with the 20 to 25 percent of electrons with energy greater than the mean value. Half the ejected electrons have energies below 10 to 15 eV - i.e. over

Summary of Data for Two Representative Low Z Gases
Bombarded by Protons Between 0.3 and 1.5 MeV

Target Gas	Proton Energy Tp, keV	Fraction of Secondary Electrons and Associated Kinetic Energy Above:								
		Mean Electron Energy $\bar{\epsilon} \int_0^\infty \sigma(\epsilon) d\epsilon = \int_0^\infty \epsilon \sigma(\epsilon) d\epsilon$			Energy Mean $\bar{\epsilon} \int_0^\infty \epsilon \sigma(\epsilon) d\epsilon = \int_0^\infty \epsilon^2 \sigma(\epsilon) d\epsilon$			Number Median $\int_0^{\epsilon_m} \sigma(\epsilon) d\epsilon = 1/2 \int_0^\infty \sigma(\epsilon) d\epsilon$		
		$\bar{\epsilon}, \text{eV}$	N	E	$\bar{\epsilon}, \text{eV}$	N	E	ϵ_m, eV	N	E
Hydrogen	300	34	0.23	0.75	160	0.044	0.37	12.5	≈ 0.5	0.91
	500	39	0.21	0.76	240	0.030	0.36	12.5		0.92
	1000	45	0.19	0.76	430	0.017	0.34	13.0		0.92
	1500	51	0.17	0.76	605	0.012	0.31	13.0		0.88
Nitrogen	300	47	0.26	0.88	168	0.062	0.38	18.0	≈ 0.5	0.92
	1000	57	0.19	0.77	430	0.026	0.31	15.0		0.93
	1400	57	0.19	0.77	530	0.016	0.30	15.0		0.94

Target Gas	Proton Energy Tp, keV	Energy Median $\int_0^{\epsilon_m} \epsilon \sigma(\epsilon) d\epsilon = 1/2 \int_0^\infty \epsilon \sigma(\epsilon) d\epsilon$			Locally deposited range $< 10^6 \text{ g/cm}^2$		
		$\bar{\epsilon}_m, \text{eV}$	N	E	ϵ, eV	N	E
Hydrogen	300	100	0.060	≈ 0.5	200	0.032	0.31
	500	136	0.062			0.040	0.40
	1000	187	0.044			0.043	0.49
	1500	225	0.040			0.047	0.53
Nitrogen	300	117	0.10	≈ 0.5	200	0.046	0.31
	1000	232	0.054			0.063	0.54
	1400	250	0.048			0.060	0.55

half of the ejected electrons are incapable of further ionization.

On the other hand, 90 to 95 percent of the kinetic energy is associated with that half of the ejected electrons having energy in excess of 10 to 15 eV. Half the kinetic energy transferred to secondary electrons is associated with fewer than 10 percent of the electrons and this percentage rapidly decreases with increasing proton energy. Electrons ejected with sufficient energy to travel distances greater than 10^{-6} g/cm² away from the proton path (i.e. kinetic energy greater than 200 eV) number only 3 to 6 percent of the total, but they transport between 30 and 60 percent of the available kinetic energy. It is these few energetic secondaries that are responsible for the large difference between the observed and calculated relative variance in the energy deposition in small volumes.

Hit Theory for Low-LET Radiation

W. C. Roesch

The time-dependent hit theory⁽³⁾ discussed before⁽⁴⁾ proved difficult to evaluate numerically in the cases of low-LET radiation such as x or gamma rays because of the small size of some of the parameters (or, equivalently, because of the large number of hits required to produce death). Two approximate expressions for the survival have now been derived for low-LET radiation. Both can be shown to be quite accurate approximations.

One approximation is made by assuming that the hits are uniformly spaced in time rather than randomly

spaced. The other is made by averaging over the random distribution in time by pairs of hits rather than averaging all of them at once. It is possible to show that the first approximation is larger than the true survival and the second, smaller. Numerical results confirm that the first approximation is always larger than the second; however, they also show that the two do not differ significantly in the final results they give for the survival. The figure shows survival curves calculated for several different dose rates (c/b) from the two approximations open and closed circles; the two sets of points essentially lie on common curves. Since the two approximations bracket the true value and since they are indistinguishable, either one can be taken as the true survival for practical purposes. The two solid lines in the figure are limiting curves for very low and very high dose rates; both of these can be derived without using the approximations.

The expression for the survival, S , for an irradiation of length t given by the second approximation is

$$S = N(1 - e^{-bt}) e^{-D/D_0} \quad (1)$$

where b is a constant and N and D_0 are what are commonly called the extrapolation number and the mean lethal dose; D is the absorbed dose. The theory yields expressions for N and D_0 in terms of more fundamental parameters.

Haynes⁽⁵⁾ and Kellerer⁽⁶⁾ have

given heuristic derivations of equations of the same form as Equation (1), and these have already been found useful in radiobiology.

The same approximate treatment yields

$$S = N(1 - e^{-bt_1})(1 - e^{-bt_2}) \quad (2)$$

$$(1 - e^{-bu}) S^*$$

for the survival to irradiation in two periods, t_1 and t_2 , separated by a time u . S^* is the survival calculated from Equation (1) for irradiation for a single period t_1+t_2 . This expression predicts correctly the basic properties found in fractionation experiments.⁽⁷⁾

Hit Fury Calculations

Theory of the Oxygen Effect

W. C. Roesch

Alper and Howard-Flanders⁽⁸⁾ summarized the data on the oxygen effect at low dose rates with the observation that the D_0 at an oxygen concentration M was related to that, D_{0N} , for $M = 0$ (i.e. in nitrogen) by

$$D_{0N}/D_0 = (KM + k)/(M + k) \quad (1)$$

where K and k are constants.

The hit theory reported above can explain these low-rate data as follows: An individual hit product can interact with the medium through reactions that either involve oxygen or through some that do not involve oxygen. Let the probability of the former be $(C/k)M/(CM/k + C)$ and the latter, $C/(CM/k + C)$, where C is a constant. (C and C/k play the role of independent constants, but they have been chosen to lead to the result of Equation (1). Similarly for s_N and Ks_N in the next sentence.) Let the probability of a reaction with oxygen leading to death be Ks_N , and one without oxygen be s_N . Then the probability of death is

$$\begin{aligned} & (C/k)MKs_N/(CM/k + C) + Cs_N/(CM/k + C) \\ & = s_N (KM + k)/(M + k) \end{aligned} \quad (2)$$

It can be shown that this expression leads to the Alper and Howard-Flanders law.

Dewey and Boag⁽⁹⁾ found that at very high dose rates the survival for low concentrations of oxygen was nearly the same as that for no oxygen. The points in the figure, taken from Epp et al.,⁽¹⁰⁾ show the phenomenon in more detail. In general, the survival at low doses is smaller than in nitrogen, but the survival curve soon turns and runs parallel to the curve for nitrogen. Dewey and Boag explained the high-rate effect by assuming that the early part of the irradiation pulse consumed the available supply of oxygen; then, during the remainder of the pulse, the population was being irradiated in anoxia. The pulses are so short that the resupply of oxygen during the pulse is negligible.

The Dewey-Boag hypothesis can be coupled with the theory above to give a quantitative explanation of the high-rate effect. It is assumed that the chance of death from an individual hit is given by Equation (2) and that M decreases from one hit to the next. The outcome then depends on the rate of decrease of M . Assuming that the rate of decrease of M is $AM/(M + B)$, where A and B are constants chosen to give the best fit to the data, leads to the solid-line curves in the figure. The chosen parameters give a good fit to the data for the higher oxygen concentrations. Further study of the consumption process is planned.

Comparison of Theory and Experiment for the Oxygen Effect

Stopping Power Partition and Mean
Energy Loss for Energetic Protons in
Hydrogen

W. E. Wilson

The total mass stopping power, $S(E_0)$, for a particle of energy E_0 can be related to the various modes for energy loss by the expression

$$S(E_0) = \frac{\eta}{\rho} \left[\sum_x E_x \sigma_x + \sum_n \int_0^{T_{\max}} \sigma_n(T) (I_n + T) dT \right], \quad (1)$$

where η and ρ are respectively the number and mass density of the absorber, σ_x is the cross section for excitation of the absorber molecules to state x , with excitation energy E_x . $\sigma_n(T)$ is the ionization cross section for ejecting a secondary electron from shell n with energy between T and $T + dT$; I_n is the binding energy of electrons in the n^{th} shell of the absorber atoms.

For incident electrons $T_{\max}$ is $(E_0 - I_n)/2$, the kinematic maximum energy that an electron of energy E_0 can transfer to an identical particle bound with energy I_n . For incident protons (or any heavy particle) $T_{\max}$ is approximately $4E_0 m_e/m_p$; $\sigma_n(T)$ is observed to decrease rapidly for T greater than this value. This limit is the classical maximum energy transfer that a particle of energy E_0 and mass m_p can give to a free particle of mass m_e .

The three terms of Equation 1 are "partial stopping powers" and are assigned the symbols

$$S_E = \frac{\eta}{\rho} \sum_x E_x \sigma_x \quad ; \quad (2A)$$

$$S_I = \frac{\eta}{\rho} \sum_n I_n \int_0^{T_{\max}} \sigma_n(T) dT = \frac{\eta}{\rho} \sum_n I_n \sigma_n \quad ; \quad (2B)$$

$$S_T = \frac{\eta}{\rho} \sum_n \int_0^{T_{\max}} \sigma_n(T) T dT = \frac{\eta}{\rho} \int_0^{T_{\max}} \sigma(T) T dT \quad ; \quad (2C)$$

σ_n is the n^{th} shell total ionization cross section, and $\sigma(T)$ the cross section for ejection of electrons with energy between T and $T + dT$ while leaving the residual ion in any state.

Recently measured cross sections for total ionization and for electron ejection have been employed to obtain the partial stopping powers defined by Equations 2A, B and C for the case of energetic protons incident upon hydrogen gas. The results indicate that the partial stopping power, S_T , which is the transfer of proton Kinetic Energy into Kinetic Energy of first generation secondary electrons, constitutes about 60% of the total stopping power from 0.2 to 1.5 MeV and is nearly independent of primary proton energy. S_I makes up about 30% of the total stopping power at 0.2 MeV and decreases to about 20% at 1.5 MeV. Experimental data are not available for the σ_x , but an estimate of S_E from theoretical calculations indicates that S_E can reasonably account for the remaining part of the total stopping power.

Energy-restricted linear collision stopping power (L_Δ) is related to the fundamental cross sections by the relation

$$L(\Delta) = \frac{n}{\rho} \left[\sum_x E_x \sigma_x + \sum_n I_n \sigma_n + \int_0^{\Delta} \sigma(T) T dT \right], \quad (3)$$

for $E_x < \Delta$ and $I_n < \Delta$ and where Δ is defined to be the energy cutoff for each energy loss channel rather than the limit on the total energy transfer in the ionizing collision.

The ICRU has defined restricted linear collision stopping power to be the quotient of dE by dl where dE is the mean energy-loss in dl due to collisions with energy transfers less than some specified value Δ . Placing the restriction on the sum of E_x , I_n and T couples the Kinetic Energy of the ionization electron with the final state of the ion and the development which leads to Equation 3 can not proceed. The definition used here leads to calculable quantities without the need for implicit assumptions or approximations.

To the extent that E_x and I_n are "locally absorbed energies" we can write (3) as

$$L(\Delta) = S_E + S_I + \int_0^{\Delta} \sigma(T) T dT; \text{ or} \quad (4)$$

$$L(\Delta) = S - S_T(\Delta), \text{ where} \quad (5)$$

$$S_T(\Delta) = \frac{n}{\rho} \int_{\Delta}^{T_{\max}} \sigma(T) T dT. \quad (6)$$

Equation 6 is a generalization of (2C). Equation 5, along with curves or tables of values for $S_T(\Delta)$ defined by Equation 6, can be used to obtain energy re-

stricted stopping powers for arbitrary cutoffs Δ . Curves of $S_T(\Delta)$ for energetic protons stopping in hydrogen were evaluated and are to be published in Radiation Research in February.

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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 experimental measurement of the doubly differential cross sections for electron ejection by monoenergetic protons, investigation of the partition and distribution of energy in the radiation field and in the irradiated medium and determination of the rates and mechanism for energy transfer in the early stages of molecular rearrangement.

Electron Emission Cross Sections

L. H. Toburen

The investigation of electron emission cross sections for a number of gases has been carried out during the past year. Particular emphasis was placed on cross sections measured for 0.3 to 1.5 MeV protons on hydrogen and for 1.0 and 2.0 MeV protons on helium. These gases are especially interesting as they are the only atoms for which theoretical calculations exist in which electron emission cross sections differential in both electron energy and emission angle have been calculated. Comparison of measured and calculated cross sections yield information regarding the details of the emission process such as the effects of long range electron-proton interactions and the effects of the distortion of the target atom by the impulsive interaction of the passing proton. In a more general sense, the range of validity of the Born approximation is tested by these results. It was found, for example, that for proton energies

as high as 1.5 MeV the Born approximation was not adequate to provide accurate double-differential cross sections for electron emission into small angles (less than 40 degrees).⁽¹⁾ Calculations by Macek based on Faddeev's equations,⁽²⁾ although only qualitatively correct for low energy proton impact, were found in excellent agreement with our measurements for 1.5 MeV protons on hydrogen. This evidence of agreement was not repeated, however, when Macek's results for helium were compared with our measurements at 1.0 MeV.⁽³⁾ This result indicates that the interactions and/or wave functions included in his calculations do not properly reflect changes in the target atom. It appears that a good deal of theoretical work will be necessary before one can accurately calculate double-differential electron emission cross sections for even the most simple atoms.

In addition to the detailed study of hydrogen and helium, considerable effort has been directed to more complex atoms and molecules. Electron

emission cross sections have been measured for a series of hydrocarbons including methane, ethane, ethylene, acetylene, and benzene in order to investigate the effects of molecular binding.

We are still in the process of analyzing results obtained from H_2^+ impact on molecular hydrogen. These data are quite complicated since we must deal with electrons ejected from the target atom by both incoming simultaneous protons and the attached electron. The incident electrons display both elastic and inelastic scattering from the target molecule.

Inner Shell Ionization and Fluorescent Yields

L. H. Toburen

Auger electron peaks are observed superimposed on the continuum electron emission from hydrocarbon targets under proton bombardment. These Auger electrons result from filling vacancies in the K-shell of carbon which are produced by proton impact. Since the fluorescent yield for carbon is small ($\omega_K \approx 0.002$) the Auger electron yield can be directly correlated with the number of inner shell vacancies produced. When different types of carbon-containing molecules were investigated, the shape and intensity of the Auger electron spectra were found to change. These changes in the shape of the Auger spectra indicate that different initial and final states are being excited for a given proton energy depending on the molecular surroundings of the carbon atom. For 1.0 MeV the total cross section for K-Auger elec-

tron emission from carbon was found to vary by 10% as the carbon-carbon bonding progressed from single to triple bonds, e.g., in going from ethane to acetylene. A variation of only 6% was found for similar measurements with 0.3 MeV incident protons. Our present electron energy resolution (3.5%) is not sufficient for detailed analysis; however, if better resolution can be obtained, information regarding final charge states of the residual target particles can be obtained, both as a function of incident particle energy and as a function of the molecular bonding.

The K-shell ionization cross sections obtained in our work are the first measured values which do not rely on an accurate knowledge of the fluorescent yield; this is significant since reported values of the fluorescent yield of carbon vary by an order of magnitude. Our results are in excellent agreement with semiclassical calculations reported by Garcia;⁽⁴⁾ calculations based on the Born approximation⁽⁵⁾ result in cross sections nearly 30% smaller.

In principle, the fluorescent yield of carbon can be obtained from our Auger electron yields and published X-ray yields.⁽⁵⁾ The fluorescent yields determined in this manner are found to be a slowly varying function of the proton energy, e.g., $\omega_K = 0.0016$ for 0.3 MeV protons and $\omega_K = 0.0025$ for 2.0 MeV proton impact. This variation of fluorescent yield with particle energy can be qualitatively explained as due to the production of multiply charged target

particles. There is presently insufficient information, however, to explain this phenomenon quantitatively. These results point out the need to redefine what is meant by the fluorescent yield to include the dependence on the final state of the residual target ion. The fluorescence efficiency appears to depend not only on the final charge state but also on the final molecular state in which the atom containing the initial inner shell ionization resides.

Our preliminary measurements for protons on xenon indicate that M- and N-shell ionization cross sections for heavy atoms may be obtained from electron emission measurements. There is, however, still some question regarding the reliability of our measurements for xenon and any conclusions await experimental work which is in progress at the present time.

Fluorescent Decay of Organic Liquids

M. L. West

Investigation of the fluorescent decay of organic liquids excited by ionizing and non-ionizing radiation has continued throughout the year. Fluorescing molecules produced under different modes of excitation serve as sensitive probes through their extreme sensitivity to the surrounding environment and allow us to explore the temporal stages of radiation action leading up to permanent molecular alterations. The non-exponential fluorescent decay previously observed in solutions of benzene in cyclohexane under proton irradiation has suggested a diffusional quenching model whereby excited molecules cre-

ated along a localized proton track interact as they diffuse from their initial reaction site. Diffusion kinetic theory as first formulated by Samuel and Magee⁽⁶⁾ to explain the effect of linear energy transfer on radical and molecular yields in water has been developed and programmed for computer manipulation. This is a convenient starting point in our effort to ultimately treat stochastically the effects of ionizing radiation in liquid solution. Since we are interested in the time behavior of molecular fluorescence as influenced by inhomogeneous radical distributions along a track, the following expression was derived:

$$\frac{d}{dt} \ln I(t) = -k_{uv} - Ck_r R(t) \quad (1)$$

where $I(t)$ is fluorescence intensity, $R(t)$ is radical concentration, k_{uv} is a rate constant from u.v. excitation, and k_r is the rate of radical-excited state interaction. Other model parameters include initial radical and excited state densities and radical-radical reaction rates. From the radiation chemist's point of view, $R(t)$ is negligible after 10^{-9} in terms of radical-radical interaction or radical scavenging and its effect on final yields. But for fluorescence decay, estimates based on approximate values of model parameters show that the last term of Equation (1) is still comparable to k_{uv} in the time interval $10^{-9} - 10^{-8}$ sec. Attempts are being made to fit this equation to experimental decay curves. Using approximate values of model parameters fixed by independent experiments in radiation chemistry we show that the

observed decay is consistent with this quenching model. Model calculations are presently being extended to include α/β ratios as an additional check on its success to explain radiation effects. Additional tests will include experiments on selected aqueous solutions where product yields and rate parameters are accurately known.

Electron-Ejection Cross Sections for Several Representative Hydrocarbon Molecules

W. E. Wilson

Cross sections, differential in ejection energy and angle, for electron emission from methane, ethane, ethylene, acetylene and benzene molecules following proton impact have been measured for protons of 0.3 and 1.0 MeV. The purpose in making the measurements is to look for an effect on the electron yield attributable to the chemical form or molecular binding of the target atoms. If such an effect is significant, it has implications for the additivity rules for ionization and energy loss in gaseous mixtures. For example, the ionization one observed in a "tissue equivalent" gas might then depend on whether methane or acetylene were used for the carbon component.

In general, the results indicate that for many practical situations involving energy transport by energetic secondaries there is no significant binding effect. The distributions differential in energy only, obtained by integrating over emission angle, have proportional zero, first, and

second moments, so that ratios of the moments are constants for the molecules studied. For all five target molecules and for both proton energies, first generation secondary electrons (FGSE) with energies greater than the mean energy, account for 74% of the total energy loss associated with Kinetic Energy of all secondaries. For both proton energies the distributions, differential in energy only, are scalable in terms of the number of outer or weakly bound electrons in the molecule. If the yield or cross section in units of $\text{cm}^2\text{eV}^{-1}\text{molecule}^{-1}$ is divided by the total number of outer electrons in the molecule (total electrons minus the carbon K-shell electrons) the result is a "universal curve" in units of $\text{cm}^2\text{eV}^{-1}\text{electron}^{-1}$. The yield "per electron" is nearly identical for all 5 molecules for secondaries with energies greater than 10 eV. Below 10 eV the data are too sparse to provide a good comparison.

This scalability is equivalent to saying the effective yield per carbon atom is the same for all 5 molecules studied. This does not say that chemical environment or molecular binding in general makes little difference in the yield of secondary electrons. We can demonstrate a definite molecular binding effect and at the same time shed some light on the validity of the Bragg-additivity rule for stopping power by using previously measured molecular hydrogen electron-emission cross sections, properly scaled, to subtract the contribution of the hydrogen from the hydrocarbon yields. The result is an equivalent carbon atom yield and the process is

the reverse of the additivity rule. When one constructs a secondary-electron energy distribution for carbon by taking $1/2$ the difference between the acetylene yield and the molecular hydrogen yield $[1/2 (C_2H_2 - H_2)]$, one obtains a result different from that obtained by subtracting from the methane distribution 2 times the hydrogen distribution, $[CH_4 - 2H_2]$. The difference shows up as a larger yield of secondary electrons for energies up to about 60 eV for the distribution synthesized from the methane-hydrogen data; the yield of 10 eV secondaries is 30% larger for the carbon atom from methane than from acetylene by this method of analysis. Distributions synthesized in a similar manner from ethane-hydrogen and ethylene-hydrogen agree more closely with the distribution obtained from methane-hydrogen whereas the distribution obtained from benzene-hydrogen falls with the distribution for the acetylene-hydrogen, i.e., the saturated hydrocarbons produce similar

secondary electron-energy distributions for carbon atoms and the unsaturated hydrocarbons indicate a different distribution.

The important conclusion we reach is that use of the Bragg-additivity rule can lead to error in predicting the number of ion-electron pairs with secondary electron Kinetic Energy as large as 60 eV.

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RADIOLOGICAL SCIENCES DEPARTMENT STAFF

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J. H. Reeves
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