Pacific Northwest Laboratory
Annual Report for 1975
to the ERDA Division of Biomedical
and Environmental Research

Part 4 Physical and Analytical Sciences
and
Analysis and Assessment

February 1976

Prepared for the U.S. Energy
Research and Development Administration
under Contract E(45-1):1830

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
Under Contract E(45-1)-1830

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$7.00; Microfiche \$2.25

3 3679 00059 2438

BNWL-2000 PT4

UC-48

PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1975 TO THE ERDA
DIVISION OF BIOMEDICAL AND ENVIRONMENTAL
RESEARCH

PART 4 PHYSICAL AND ANALYTICAL SCIENCES
AND
ANALYSIS AND ASSESSMENT

By
J. M. Nielsen
and Staff Members of
Radiological Sciences Department
Physics and Instrumentation Department
Systems Department

February 1976

Battelle
Pacific Northwest Laboratories
Richland, Washington 99352

PREFACE

The Annual Report for 1975 to the U.S. Energy Research and Development Administration's Division of Biomedical and Environmental Research is organized by major categories according to our schedule-189 submissions. Each part is directed toward a particular DBER Research and Development Program: Part 1 to Biomedical Programs, Parts 2 and 3 to Environmental Programs, and Part 4 to Physical and Technological Programs. Each part of the Annual Report comprises project reports authored by scientists from several research departments, reflecting the interdisciplinary nature of the research effort. The Annual Report consists of four parts:

Part 1	Biomedical Sciences		
	Program Manager - W. R. Wiley	R. C. Thompson, Report Coordinator	D. L. Felton, Editor
Part 2	Ecological Sciences		
	Program Manager - B. E. Vaughan	B. E. Vaughan, Report Coordinator	J. L. Helbling, Editor
Part 3	Atmospheric Sciences		
	Program Manager - C. L. Simpson	C. E. Elderkin, Report Coordinator	E. L. Owzarski, Editor
Part 4	Physical and Analytical Sciences		
	Program Manager - J. M. Nielsen	J. M. Nielsen, Report Coordinator	G. M. Garnant, Editor
	Analysis and Assessment		
	Program Manager - J. C. Fox	J. M. Nielsen, Report Coordinator	G. M. Garnant, Editor

Reports previously issued are as follows:

Annual Report for

1951	HW-25021, HW-25709
1952	HW-27814, HW-28636
1953	HW-30437, HW-30464
1954	HW-30306, HW-33128, HW-35905, HW-35917
1955	HW-39558, HW-41315, HW-41500
1956	HW-47500
1957	HW-53500
1958	HW-59500
1959	HW-63824, HW-65500
1960	HW-69500, HW-70050
1961	HW-72500, HW-73337
1962	HW-76000, HW-77609
1963	HW-80500, HW-81746
1964	BNWL-122
1965	BNWL-280, BNWL-235, Vol. 1-4
1966	BNWL-480, Vol. 1, BNWL-481, Vol. 2, Pt. 1-4
1967	BNWL-714, Vol. 1, BNWL-715, Vol. 2, Pt. 1-4
1968	BNWL-1050, Vol. 1, Pt. 1-2, BNWL-1051, Vol. 2, Pt. 1-3
1969	BNWL-1306, Vol. 1, Pt. 1-2, BNWL-1307, Vol. 2, Pt. 1-3
1970	BNWL-1550, Vol. 1, Pt. 1-2, BNWL-1551, Vol. 2, Pt. 1-2
1971	BNWL-1650, Vol. 1, Pt. 1-2, BNWL-1651, Vol. 2, Pt. 1-2
1972	BNWL-1750, Vol. 1, Pt. 1-2, BNWL-1751, Vol. 2, Pt. 1-2
1973	BNWL-1850, Pt. 1-4
1974	BNWL-1950, Pt. 1-4
1975	BNWL-2000, Pt. 1-4

W. J. Bair
 Manager, Biomedical and Environmental
 Research Program

FOREWORD

Part 4 of the Pacific Northwest Laboratories Annual Report for 1975 to the ERDA Division of Biomedical and Environmental Research continues the same format as for 1974 and includes those programs funded under the title Physical and Analytical Studies. Included are the programs in Instrumentation Technology which cover radiological and chemical methods and instrument development and studies; programs in Radiological and Chemical Physics which include radiation physics, radiation biophysics and radiation dosimetric research; Analytical and Evaluational Studies of nuclear fuel components, social value assessments, and statistical and epidemiological studies of environmental control; and a Regional Studies Program for assessment of effects from energy development and conservation.

CONTENTS

PREFACE	ii
FOREWORD	iii
INSTRUMENTATION TECHNOLOGY	
Radiation Instrumentation--Radiological Chemistry	1
Low-Level Radiological Chemistry Laboratory Techniques	2
Comparison of Ge(Li) Diode Efficiencies for Two Counting Geometries - N. A. Wogman and J. C. Laul	2
Detection Sensitivity Limitations for the Determination of the ^{232}U Daughters - N. A. Wogman	3
Background Reduction in Conventional Ge(Li) Gamma-Ray Spectrometers - N. A. Wogman	5
Specialized Utilization of Multidimensional Gamma-Ray Spectrometer Systems - N. A. Wogman	6
The Determination of ^{241}Am and ^{244}Cm Using an Intrinsic Germanium Detector - C. W. Thomas	8
Instrumentation Research and Development - J. R. Kosorok	12
Measurement of Ultra Trace Quantities of Several Elements in CTR Experiments - J. C. Langford	13
In Situ Monitoring	14
In Situ Measurement of Plutonium and Americium in Soils - K. K. Nielson, N. A. Wogman and R. L. Brodzinski	14
Radium Accumulation in Animal Thyroid Glands...A Possible Method for Uranium and Thorium Prospecting - N. A. Wogman and R. L. Brodzinski	16
Environmental Pollution Analysis.	18
Analysis of Arsenic in Solution by X-Ray Fluorescence of Ferric Hydroxide Precipitate - E. A. Crecelius	18
An Improved Collimator for X-Ray Fluorescence Analysis - K. K. Nielson and N. W. Wogman	18
Trace Element Analysis of Natural Waters Using Energy Dispersive X-Ray Fluorescence - K. K. Nielson	20
Corrections for X-Ray Absorption in Aerosol Samples Using X-Ray Scattering - K. K. Nielson	21
Application of Mercury Speciation Traps for Geothermal Gases - E. A. Crecelius	22

Analysis of Trace Elements in Sea Water by Anodic Stripping Voltametry - E. A. Crecelius and J. C Kutt	23
In Situ Pollutant Measurements	24
In Situ X-Ray Fluorescence Analyzer Improvements - N. A. Wogman	24
An Improved Method for Automatic Peak Analysis in X-Ray Spectra - K. K. Nielson	26
Trace Constituent Analysis by Laser Excitation	28
The Analysis of ^{85}Kr by Laser Excitation Techniques - B. A. Bushaw and T. J. Whitaker	28
Analytical Techniques for Measurement of ^{99}Tc in Environmental Samples	31
Ultrasensitive Measurement of ^{99}Tc by Mass Spectrometry - J. H. Kaye	31
Fast Real-Time Measurement of Extremely Low-Level Airborne Plutonium	33
Fast Real-Time Measurement of Low-Level Airborne Plutonium - J. J. Stoffels and C. R. Lagergren.	33
Environmental Application of Holography	36
Application of Holography to Environmental Studies - B. P. Hildebrand	36
Chemical Reaction Kinetics	38
Measurement of Combustion Products by Chemical Reaction Kinetics - J. A. Campbell, W. D. Felix and N. A. Wogman	38
References	41
Publications and Presentations	42
RADIOLOGICAL AND CHEMICAL PHYSICS	
Radiation Physics	45
Electron Emission from Proton Bombardment of Molecular Targets - L. H. Toburen, W. E. Wilson, T. E. Porter, and D. J. Lynch	46
Electron Emission from Proton Bombardment of Carbon Foils - L. H. Toburen, H. Paretzke, and W. E. Wilson	48
Consistency Tests of the Measured Cross Sections - L. H. Toburen and S. T. Manson	49
Ionization of Heavy Ion Collisions - L. H. Toburen and W. E. Wilson	53
Pulsed Radioluminescence Studies - J. H. Miller and M. L. West	55

Radiation Dosimetry and Radiation Biophysics.	62
Division Delay After Low X-Ray Doses and the Effect of Cycloheximide - M. H. Schneiderman, L. A. Braby, and W. C. Roesch	62
The Mitotic Selection Technique - M. H. Schneiderman	66
Hydroxyurea: A G ₂ Effect - M. H. Schneiderman, D. V. Leeper, and W. C. Dewey	66
Dose Rate Effects and <i>Chlamydomonas</i> - L. A. Braby and W. C. Roesch	67
Analytical Expressions for Cross Section Data - W. E. Wilson and L. H. Toburen	69
Microdosimetry of Internal Sources - W. C. Roesch	72
Experimental Microdosimetry for Alpha Particles - L. A. Braby and W. C. Roesch	73
Dosimetry of Particulate Sources in the Lung	76
Calculations for Microdosimetry of Internal Sources - W. C. Roesch	76
References	79
Publications and Presentations	81
HEALTH PROTECTION	
Dosimetry of Internal Emitters	83
Dose to G.I. Tract from Ingested Insoluble Beta Emitters - F. T. Cross, G. W. R. Endres, F. N. Eichner, R. A. Libby and M. F. Sullivan	83
ANALYSIS AND EVALUATION	
Toxic Substances in Nuclear Fuels	87
Toxic Effluents from UO ₂ Fuel Cycle - R. M. Fleischman, W. A. Muckerheide, K. J. Schneider and R. L. McKeever	87
Assessment of Social Values in Thermal Plant Siting	89
Social Values in Thermal Plant Siting - J. B. Burnham, J. C. King and J. A. Hébert	89
Considerations and Methodologies for Studies Concerning the Effect of Low-Level Exposures to Environmental Contaminants	91
Statistical Considerations of the Effect of Low-Level Exposures to Environmental Contaminants - E. S. Gilbert, R. L. Buschbom, and E. B. Perrin	91
References	92
Publications and Presentation	93

REGIONAL STUDIES

Pacific Northwest Regional Assessment Program	95
Energy--Economic Sectors - J. W. Currie, L. E. Erickson, D. W. Fraley, T. P. Harrington	97
Demographic Sector - J. J. Jacobsen	102
Air Resources Sector - D. S. Renne and D. L. Elliott	104
Land and Water Resource Sectors - M. W. Stradley and S. J. Shupe	106
Social Sector - M. E. Olsen	108
Psychological Sector - D. H. Summers	110
Bioproductivity Sector - R. H. Sauer	110
Institutional Constraints to Energy Development - M. Curry and M. Greene	112
Special Assessments - W. H. Swift, and G. L. Wilfert	112
References	119
PUBLICATIONS AND PRESENTATIONS	123
ORGANIZATION CHARTS	127

INSTRUMENTATION TECHNOLOGY

- **RADIATION INSTRUMENTATION—RADIOLOGICAL CHEMISTRY**
- **ENVIRONMENTAL POLLUTION ANALYSIS**
- **IN SITU POLLUTANT MEASUREMENTS**
- **TRACE CONSTITUENT ANALYSIS BY LASER EXCITATION**
- **ANALYTICAL TECHNIQUES FOR MEASUREMENT OF ^{99}Tc IN ENVIRONMENTAL SAMPLES**
- **FAST REAL-TIME MEASUREMENT OF EXTREMELY LOW-LEVEL AIRBORNE PLUTONIUM**
- **ENVIRONMENTAL APPLICATION OF HOLOGRAPHY**
- **CHEMICAL REACTION KINETICS**

- **RADIATION INSTRUMENTATION—RADIOLOGICAL CHEMISTRY**

The development and utilization of radioanalytical technology and instrumentation allows studies of the environmental behavior of trace amounts of radionuclides and their stable element counterparts. During the past year, emphasis has been placed on improving the sensitivity of radioanalytical analysis systems and techniques as well as feasibility studies which focus the future direction of the program.

Low-Level Radiological Chemistry Laboratory Techniques

In Situ Monitoring

Low-Level Radiological Chemistry Laboratory Techniques

Newly developed electronic and computer technology together with improved low-background construction materials and techniques have aided in developing more sensitive radiation instrumentation. Backgrounds have been reduced by 10-fold at the Th and U photopeak areas, as well as 20- to 100-fold in the ^{40}K photopeak region for NaI(Tl) spectrometers and from 5-fold for Th and U to 30-fold for ^{40}K for Ge(Li) spectrometers. High sensitivity x-ray analysis with intrinsic Ge, coupled with computer techniques, allowed measurements of specific tracers for resuspension studies and the determination of ^{241}Am and ^{244}Cm . Techniques were also developed to measure the neutron erosion of CTR construction materials.

Comparison of Ge(Li) Diode Efficiencies for Two Counting Geometries

N. A. Wogman and J. C. Laul

The direct measurement of radionuclides in environmental samples together with the analysis of activation products formed by neutron irradiation requires measurement on Ge(Li) diode spectrometers. In these measurements, samples of various sizes must be analyzed and it is important to know how efficiencies and

resolutions vary with sample size and sample location relative to the diode. In this work we have compared counting efficiencies and also examined resolutions for samples as a function of vertical and horizontal distance from an "uplooking" diode. The coaxial diode used in this study was 54.5 mm in diameter by 51.8 mm long with a drift depth of 20.7 mm. The distance from the face of the diode to the face of its vacuum can was about 2 mm. In Figure 1.1 we have

FIGURE 1.1. Relative Counting Efficiency of ^{60}Co as a Function of Vertical and Horizontal Distance to a 25% Efficient Ge(Li) Diode

plotted the relative counting efficiency of the 1.33 MeV photopeak of ^{60}Co as a function of distance from the source for both vertical and horizontal separation. These efficiency curves are for a 1.3 cm x 5 cm long liquid source and lie within 15% of each other over the source to detector separation distances of interest. The resolution for horizontal versus vertical source-to-detector separation distances varies by less than 0.1 KeV, and the peak-to-Compton ratios are within 5%. Further measurements as a function of thickness for 1 cm diameter ^{137}Cs liquid samples in going from 2 to 14 mm in thickness showed less change in efficiency for side mounting versus top mounting configurations. The use of a Marinelli type beaker for liquid samples proved justified and advantageous in providing optimum detector responses.

Detection Sensitivity Limitations for the Determination of the ^{232}U Daughters

N. A. Wogman

Studies of the deposition in organisms of radionuclides from ^{232}U , one of the major radiation hazards from recycled breeder reactor fuels, involve measurements of all their daughter products at various sites within the organism. Although some of the daughters have half lives of less than seconds, these may require measurement since a direct measure of the longer-lived daughters is impossible due to gamma-ray interference from other radionuclides. These measurements can now be made with an anticoincidence shielded Ge(Li) diode at from 1 to 10 pCi sensitivities for most of the gamma-ray emitting daughters.

The NaI(Tl) anticoincidence shielded diode utilized for these measurements has an efficiency of 15.0% and a resolution of 2.2 keV full width half maximum. The system provides a peak-to-Compton ratio of greater than 1000 to 1 for a point source, and greater than 750 to 1 for a 1-cm thick by 5-cm diameter sample. Table 1.1 shows the possible contaminants which might interfere with the direct gamma-ray analysis of the

daughter products of ^{232}U . These contaminants may be from natural contamination by the organism's uptake of natU and ^{232}Th , produced by a variety of neutron or proton reactions occurring during ^{232}U tracer production, or by neutron reactions on the fuels of a breeder reactor. With the use of the anticoincidence shielded diode, these types of contaminants do not interfere in general with the measurement of ^{232}U , or its daughters at the levels shown in Table 1.2. The daughters ^{216}Po and ^{212}mPo cannot be analyzed by this technique. Polonium-212m at 570 keV is significantly interfered with by ^{214}Po , the daughter of ^{238}U . Polonium-216 does not have gamma-rays of significant intensity to be measurable above normal background levels in even an anticoincidence shielded Ge(Li) diode system.

Table 1.2 shows that ^{232}U , ^{228}Th , ^{224}Ra , ^{220}Rn , ^{212}Pb , ^{212}Bi and ^{208}Tl can be detected by direct gamma-ray analysis of gram quantities of biological tissues containing from 1 to 30 pCi using the listed gamma-ray energies.

Since measurements are made of many individual daughter radionuclides, this technique eliminates the need for a variety of assumptions on the degree of decay equilibrium achieved within the entire organisms for ^{232}U and its daughters. As stated earlier, the identification of short half-life daughters allows the identification of the longer-lived species in the presence of contamination radioactivity. This allows knowledge to be obtained of the radiation dose to each organ and the actual distribution of radionuclide species.

Lower limits of detection could be achieved if chemistry and other counting techniques were utilized; however, for the large number of samples normally involved in a biological radiation dose study--from 1 to 10,000--the cost per sample of chemical-separation analysis is normally from \$100 to \$300 each. Thus the direct counting technique, which costs on the order of \$20 to \$50 per sample, is the most economic method for the determination of the radionuclides in these studies.

TABLE 1.1. Possible Gamma Ray Radionuclides Interfering in the Direct Measurement of ^{232}Th , ^{232}U , and Their Daughters

Isotope	γ Energy (keV)	%	Possible Contaminant ^(a) Interference	γ Energy (keV)
^{232}U	57.60	0.21	$^{233}\text{U}/\text{nth}^{232}\text{Th}$	54.69
			$^{237}\text{Np}/\text{nfa}^{238}\text{U}$	57.11
			^{232}Th NAT	59.00
			$^{237}\text{Pu}/\text{CHA}^{235}\text{U}^{237}\text{NP}$	59.54
			$^{241}\text{Am}/\text{nth}^{240}\text{Pu}$	59.54
^{228}Th	84.50	1.6	$^{211}\text{Rn}/\text{CHA}^{232}\text{Th}$	81.52
			$^{222}\text{Ra}/\text{CHA}^{232}\text{Th}^{222}\text{Pa}$	81.87
			^{226}Ra , NAT ^{238}U	81.87
			NAT ^{233}U	83.23, 86.11
			^{225}Ac , $^{229}\text{Pa}/^{229}\text{Th}$	85.50, 87.50
			$^{224}\text{Ac}/\text{CHA}^{232}\text{Th}$	83.50
			^{223}Ra , $\text{Pa}/^{227}$, NAT ^{235}U	83.78
			^{226}Ra , NAT ^{238}U	83.78
			^{222}Ra , $\text{CHA}^{232}\text{Th}$	83.78
			^{231}U , $\text{CHA}^{232}\text{Th}$	84.18
			^{231}Th , NAT ^{235}U	84.24
			^{214}Pb , NAT ^{238}U	84.80
			^{206}Bi , $\text{CHA}^{232}\text{Th}$	84.80
			^{209}Tl , NAT ^{233}U	84.80
			^{226}Ra , $\text{CHA}^{220}\text{Ra}$	85.44
			^{237}Np , nfa^{238}U	86.49
			^{233}Th , $\text{nth}^{232}\text{Th}$	86.50
			^{212}Pb	87.20
			^{206}Bi , $\text{CHA}^{232}\text{Th}$	87.30
^{224}Ra	241.0	4.2	^{212}Pb	238.63
			^{214}Rb , NAT ^{238}U	241.92
			^{226}Th , $\text{CHA}^{232}\text{Th}$	242.40
^{220}Rn	542	0.07	^{209}Tl , NAT ^{233}U	544
^{216}Po				
^{212}Pb	238.63	43.1	^{224}Ra	241.0
			^{227}Th , NAT ^{235}U	236.0
^{212}Bi	727.17	6.5	^{230}Pa , $\text{CHA}^{232}\text{Th}$	728.16
^{212}mPo	570	2.0	$^{234}\text{Pa}^{238}\text{U}$	568.70
			$^{234}\text{Pa}^{238}\text{U}$	569.50
			^{205}Bi , $\text{CHA}^{232}\text{Th}$	570.58
			^{230}Pa , $\text{CHA}^{232}\text{Th}$	571.08
^{208}Tl	583.14	86		

a. nth = thermal neutron reactions; nfa = fast neutron reactions;
 NAT = natural radioactivity; CHA = charged particle reactions
 on isotope.

TABLE 1.2. Detector Sensitivities for ^{232}Th , ^{232}U , and Their Daughters by Direct Ge(Li)(a) Gamma Ray Analysis

Radionuclide	E_γ , keV	pc ^(b)
^{232}U	57.60	30
^{228}Th	84.50	10
^{224}Ra	241	10
^{220}Rn	542	1000
^{216}Po	-	-
^{212}Pb	238.63	1
^{212}Bi	727.17	5
^{212}mPo	570	-
^{208}Tl	583.14	5

- a. An NaI(Tl) anticoincidence shielded 15% Ge(Li) diode with 2.2 keV resolution.
b. 300 min. analysis.

Background Reduction in Conventional Ge(Li) Gamma-Ray Spectrometers
N. A. Wogman

A high efficiency, low background Ge(Li) gamma-ray detector system has been constructed without anticoinci-

dence shielding which provides a ^{40}K , ^{232}Th and ^{238}U background lower than that obtained with some specialized anticoincidence shielded Ge(Li) systems.

The new system, as shown in Figure 1.2 was assembled from a 24.9% efficiency, 1.81 keV resolution Ge(Li) detector. The cryostat-detector configuration provides a 90° angle between the detector and the cryostat cold finger which houses the molecular sieve. Thus, the molecular sieve which contains 4.4 dpm ^{232}Th , 3.0 dpm ^{238}U , and 9.0 dpm ^{40}K per gram does not have a direct line of sight to the diode when it is placed in its lead shield. The diode housing (constructed of 304 stainless steel) did not contain the often found trace impurity cobalt-60, and contained less than 0.006 dpm ^{40}K , ^{238}U and ^{232}Th per gram of steel. Uranium and thorium daughters were detected in the preamp assembly which was therefore shielded from the diode with lead-iron shield. Future assembly configurations designed for low background should place this preamp type near the liquid nitrogen cryostat, protected by a 90° angle from the diode. The shield consisted of a 15-cm lead cave lined with 5-cm thick pre-World War II steel. The entrance hole

FIGURE 1.2. High Efficiency-Low Background Ge(Li) Gamma-Ray Detector System

through which the Ge(Li) diode penetrated the standard shield was shadow-shielded from external radiation sources with 10 cm of lead as shown in Figure 1.2.

As is shown in Table 1.3, the ^{208}Tl (^{232}Th daughter) background decreased from 0.56 cpm with 5 cm of iron shielding to 0.08 cpm inside the 5-cm Fe + 15-cm Pb + 10-cm lead shadow-shielded cave. This same cave decreased the ^{214}Bi background from 0.41 to 0.02 cpm while the ^{40}K peak area decreased from 5.1 cpm to 0.015 cpm. The use of iron as the interior shield eliminates the necessity of using a cadmium-copper interior to reduce the lead x ray produced by radiation interaction with normal lead caves. The specially shielded Ge(Li) detector system described here provides a background of 0.2 cpm in the 75 keV Pb x-ray region. A similarly sized uplooking diode in a 10-cm lead cave has a ^{208}Tl background of 0.2 cpm, a ^{214}Bi background of 0.05 and a ^{40}K background of 3.5 cpm. This latter diode when placed in a NaI(Tl) crystal anticoincidence shield still showed a background of 0.02 and 0.1 cpm ^{208}Tl in the noncoincident and coincident backgrounds, respectively. Bismuth-214 backgrounds for this shielded diode were 0.01 and 0.02 cpm in the noncoincident and coincident areas, respectively; and ^{40}K was 0.03 and 0.004 dpm in the coincident and noncoincident mode of operation. A comparison of this data with that ob-

tained from the new diode cryostat design illustrates that the new system has a background lower than the anticoincidence shielded system for both coincident and noncoincident gamma ray emitters. Of course the coincidence gamma-ray emitters are found in the coincidence spectra and the single gamma-ray emitters are detected in the noncoincident spectra. The construction cost of the new design was only \$1000 more than that of a standard uplooking diode assembly. The use of this system is presently being expanded in neutron activation analysis programs and low level environmental radionuclide analysis in ERDA-DBER programs.

Specialized Utilization of Multi-dimensional Gamma-Ray Spectrometer Systems*

N. A. Wogman

ERDA-developed multidimensional gamma-ray spectrometer systems have been used during the past year to assist in the development of basic information on remaining background in low background counting systems, and on basic decay processes. In the first, low background materials were confirmed for the antineutrino detector, Aida III developed by the Savannah River antineutrino facility. In the second program, radioisotopically

* Non-DBER program of direct relevance.

TABLE 1.3. ^{238}U , ^{232}Th , and ^{40}K Gamma Ray Backgrounds for a Large Ge(Li) Diode^(a) Under Varying Shielding Conditions

Shielding Conditions	cpm/peak area		
	^{208}Tl (^{232}Th) 584 keV	^{214}Bi (^{238}U) 1121 keV	^{40}K 1462 keV
5 cm Fe	0.56	0.41	5.1
5 cm Fe + 5 cm Pb	0.23	0.079	0.83
5 cm Fe + 10 cm Pb	0.18	0.054	0.56
5 cm Fe + 15 cm Pb	0.13	0.038	0.28
5 cm Fe + 15 cm Pb + 10 cm Shadow Shield	0.081	0.019	0.015

a. Efficiency of 24.9%, Resolution of 1.81 keV, 90° angle between diode and molecular sieve.

uncontaminated ^{82}Se material was assured for an experimental investigation of lepton conservation in the double beta decay of ^{82}Se for the Department of Physics, Columbia University.

Operation of antineutrino detectors, Aida I and II, at Savannah River Laboratory was hampered by natural background interferences leading to required modifications for background reduction. For this program, a variety of low background gamma-ray spectrometer construction materials (such as scintillators, cladding and shielding) were studied as potential substitutes for materials used previously.

As is shown in Figure 1.3 the overall background of the new Aida III system was reduced (with the low background materials) about a factor of 10 in the gamma-ray energy range from 0 to 3 MeV. The primary emphasis was to eliminate all sources of ^{40}K , ^{232}Th , ^{238}U and ^{60}Co in the new system.

Aida III is a complex detector system combining plastic phosphors, NaI(Tl) crystals, liquid scintillators and mass shielding with a variety of coincidence-anticoincidence operations possible. As a side benefit, the Aida III system with its low background could be used to identify possible sources of background by observing its response to ^{40}K , ^{232}Th and ^{238}U standards at various positions within the detector assembly. Shown in Figure 1.4 are background spectra of one of our multidimensional gamma-ray spectrometer (MDGRS) as measured with a 10 cm lead cave in an area of Grand Coulee Dam which provided 110 m of concrete overburden. The MDGRS detector system consisted of two 23-cm by 20-cm thick NaI(Tl) crystals surrounded by 38-cm by 28-cm thick NaI(Tl) anticoincidence shields. The data obtained from Aida III allowed the sources of background peaks for gamma-rays between 0 and 10 MeV in NaI(Tl) systems to be further confirmed and identified as follows (The data is shown in Table 1.4 and Figure 1.4):

1. 1.46 MeV gamma peak due to ^{40}K ,
2. 2.2 MeV gamma due to cosmic-ray neutron capture by hydrogen in the system,

3. 2.61 and 0.58 MeV gammas are from ^{208}Tl , the daughter of ^{232}Th ,
4. 2.6 MeV peak area is also from 4 MeV alpha energy deposition in the crystal due to ^{238}U ,
5. 3.2 MeV peak area is from 4.7 MeV alpha deposition in the crystal and the ^{208}Tl gamma sum energies,
6. 4.4 MeV peak area is from 6 MeV alpha of ^{218}Po in the crystal,
7. 5.7 MeV peak area is from 7.7 MeV alpha of ^{214}Po ,
8. 6.6 MeV peak area is from cosmic-ray neutron capture by ^{127}I ,
9. 7.2 MeV peak area is from alpha and beta of ^{212}Po , and
10. higher energy gammas whose sources are as yet undefined.

FIGURE 1.3. Background as a Function of Gamma-Ray Energy for the Antineutrino Detection System AIDA I, II, and III

FIGURE 1.4. Sample Crystal Background of a Multidimensional Gamma-Ray Spectrometer as a Function of Energy at Ground Level and with 110 m of High-Density Concrete Overburden

In the second study, that of lepton conservation in the double beta decay of ^{82}Se , multidimensional gamma-ray spectrometry allowed analysis of radioisotopes in high purity ^{82}Se . Very high purity was necessary since the analysis of double beta decay was dependent on the measurement of the sum energy of the two emitted electrons. The specialized use of the multidimensional gamma-ray spectrometer system allowed a lower limit to be determined for gamma-ray producing

species in the ^{82}Se source. A lower limit could then be set on the half life for double beta decay of ^{82}Se which was greater than 3.1×10^{21} yr at the 68% confidence level. Through the use of the reported lifetimes of ^{82}Se , this allowed the investigators at Columbia University to set an upper limit of 9% on the branching ratio for neutrinoless decay.

The Determination of ^{241}Am and ^{244}Cm Using an Intrinsic Germanium Detector C. W. Thomas

With the use of high exposure fuels for energy generation, the production of transuranium elements through the end of the century will be substantial. It has been estimated that about 2×10^9 curies of transuranium elements will be produced and unlike nuclear weapons, debris will include much larger amounts of ^{241}Am and ^{244}Cm than of ^{239}Pu . Consequently, there will be a need for methods to determine the concentration of both ^{241}Am and ^{244}Cm in environmental samples. To aid in measuring these isotopes, an instrumental method was developed so that these radionuclides could be measured in the presence of each other.

The high resolution, low background and low noise level of thin, large-area intrinsic germanium diodes makes this type of detector very attractive for measuring gamma rays and x rays in the 5 to 100 keV range. Figure 1.5 shows the resolution of a 400 mm² x 5 mm intrinsic diode as a function of energy. The resolution (FWHM) of this detector measured at 5.90 keV was 0.360 keV, at 59.5 keV was 0.430 keV, and at 122.1 keV was 0.550 keV. The absolute efficiency as shown in Figure 1.6 varied from 32% at 5.9 keV to 9.4% at 122 keV.

The decay characteristics of ^{241}Am and ^{244}Cm are shown in Table 1.5. Both ^{241}Am and ^{244}Cm are alpha emitters and also emit characteristic L x rays; in addition, ^{241}Am emits a gamma ray of 59.5 keV. Spectra in Figure 1.7 show the spectra obtained using equal activities of ^{241}Am and ^{244}Cm . The separation of the L x rays, especially in the L_{β} region, coupled with the 59.5 keV gamma ray of ^{241}Am , affords an excellent method to measure these isotopes in the presence of each other.

TABLE 1.4. Sources of Background for a NaI(Tl)
Multidimensional Gamma-Ray Spectrometer

Peak	Isotope/Particle	Apparent MeV	Energy Actual MeV	Efficiency of Scintillator	Notes
A & A'	$^{40}\text{K } \gamma$	1.46	1.46	1	
B & B'	Neutron Capture, $\text{H}^2 \gamma$	2.12	2.12	1	
C & C'	$^{208}\text{Tl } \gamma$	2.61	2.61	1	Illustrate a small amount of ^{208}Tl gammas adding to an equal height "Doublet."
D & D'	$^{208}\text{Tl } \gamma$	2.61 & 0.58	2.61 & 0.58	1	
E	$^{238}\text{U } \alpha$	2.6	4.15(25%) 4.20(75%)	Apx. 0.62	These emitters cause the equal height "Doublet."
F	$^{234}\text{U } \alpha$	3.2	4.77(72%) 4.72(28%)	0.67	
G not visible	$^{226}\text{Ra } \alpha$	Covered by $^{234}\text{U } \alpha$	4.78	-	
H	$^{210}\text{Po } \alpha$	Part of shoulder	5.30	-	
I	$^{222}\text{Rn } \alpha$	Shoulder Apx. 3.7	5.49	Apx. 0.67	
J	$^{218}\text{Po } \alpha$	4.14	6.00	0.69	
K					
L	$^{214}\text{Po } \alpha$	5.67	7.69	0.73	Reasonable that efficiency is higher for these alphas, since velocity is higher & ionization should be less dense.
M	$n + ^{128}\text{I } \gamma$	6.62	6.7 8.78 + apx 1.1	1	
N	$^{212}\text{Po } \alpha + \beta$	7.17	Alpha - Ave Beta	Apx. 0.72	Reasonable that efficiency is higher for these alphas, since velocity is higher & ionization should be less dense.
	$n + ^{128}\text{I } \gamma$	6.62	6.7	1	Neutron capture gammas.
	$n + ^{128}\text{I } \gamma$		6.3	1	
	$n + ^{128}\text{I } \gamma$		5.74	1	
N'	$^{212}\text{Po } \alpha$	Covered	8.78	~0.73	
O'	$n + \text{Pb } \gamma$	7.4	7.39	1	
P'	-	8.2	-	-	Fission gammas??
Q'	-	8.7	-	-	
R'	-	9.7	-	-	

For measurements the intrinsic diode was mounted in a plexiglass sample holder which in turn was housed in a 4-in. thick lead shield lined with iron plate. Background counting rates in the L_β x-ray region for ^{241}Am and ^{244}Cm and the 59.5 keV gamma ray of ^{241}Am were 0.0378 cpm, 0.0301 cpm, and 0.0272 cpm, respectively. The measurement for ^{241}Am is most easily made using the 59.5 keV gamma ray while the measurement for ^{244}Cm is made using the L_β x ray.

Interference from ^{241}Am in the L_β x-ray region of ^{244}Cm is 0.0105 times the counting rate of ^{241}Am in the 59.5 keV gamma-ray region for equal activities of the two isotopes. A common method of reporting detection limits is the value of 3 sigma of the background. Using this standard the detection levels based on a 1000-min counting period for ^{241}Am was 0.33 dpm while for ^{244}Cm the detection limit was 1.7 dpm.

FIGURE 1.5. Resolution of Intrinsic Germanium Diode as Measured by ^{55}Fe , ^{241}Am , ^{57}Co

FIGURE 1.6. Absolute Efficiency of Intrinsic Ge Diode (400 mm² x 5 mm, with 5 mil Be window)

TABLE 1.5. Decay Characteristics of ^{241}Am and ^{244}Cm

Isotope	2 1/2 (Years)	Radiation	Energy	Abundance
^{241}Am	464	Alpha	5.477 MeV	85%
		Alpha	5.435 MeV	12.6%
		Alpha	5.378 MeV	1.7%
		Alpha	Others	<1%
		L_{α} X ray	14.0 keV	$13.5 \times 10^{-2}/\alpha$ decay
		L_{β} X ray	17.8 keV	$18.4 \times 10^{-2}/\alpha$ decay
		L_{γ} X ray	20.8 keV	$5.0 \times 10^{-2}/\alpha$ decay
		Gamma	26.4 keV	$2.5 \times 10^{-2}/\alpha$ decay
		Gamma	43.0 keV	$6.0 \times 10^{-4}/\alpha$ decay
		Gamma	59.6 keV	$35.9 \times 10^{-2}/\alpha$ decay
^{244}Cm	17.9	Alpha	5.808 MeV	76.7%
		Alpha	5.766 MeV	23.3%
		Alpha	Others	<1%
		L_{α} X ray	14.3 keV	$4.0 \times 10^{-2}/\alpha$ decay
		L_{β} X ray	18.3 keV	$4.6 \times 10^{-2}/\alpha$ decay
		L_{γ} X ray	21.4 keV	$1.3 \times 10^{-2}/\alpha$ decay
		Gamma	42.9 keV	$6.2 \times 10^{-4}/\alpha$ decay
		Gamma	98.9 keV	$9.8 \times 10^{-5}/\alpha$ decay

FIGURE 1.7. Spectra of ^{241}Am and ^{244}Cm from Equivalent Activities Using Intrinsic Ge Diode

Instrumentation Research and Development

J. R. Kosorok

Instrumentation research and development areas emphasized this past year were improving the performance of hard-wired instrument systems and applying programmable controllers. These were accomplished by evaluating: 1) NIM modules in research configurations to determine their capabilities and limitations; 2) efficient use of programmable controllers for instrumentation applications; and 3) structured programming techniques.

A. Commercial NIM modules designed to reject pile-up pulses and correct for dead-time losses in single dimensional gamma-ray spectrum analyzers were evaluated. The ability to process at high count rates allows faster data collection times to reduce analysis expense and permit greater leniency in sample preparation. A typical analyzer system consisting of a Ge(Li) detector with integral pre-amp, a linear amplifier, an ADC and memory exhibits increasing gain-shift and resolution degradation as count rates increase over 1000 counts per second. The exact count rate at which errors become greater than 2%, for example, depends on factors such as the shaping time constant in the amplifier. At present, there is only one commercially available pile-up rejector and dead-time corrector that can be installed in standard analyzer systems with no special modifications to the ADC. Our evaluations have shown that systems with this module installed perform consistently with less than 2% change in spectrum peak areas over a range of 1,000 to 50,000 counts per sec. However, even this module cannot be used below a count rate of a few hundred counts per sec., or the dead-time correction fails and elapsed time intervals are incorrectly measured. An operational procedure has been developed based on the dead-time meter reading that requires the rejector-corrector to be switched out of the system at dead-times below approximately 5%. The actual dead-time depends on the amplifier time constant used with a particular system.

B. A multiple detector alpha counting analyzer was developed using commercially available components to allow simultaneous accumulation of up to eight alpha spectra. The commercial components switch only one linear signal into the main amplifier at a time, so the noise level is from an individual detector rather than from all the detectors in the system. Digital offset is provided in the ADC rather than with a biased amplifier, which eliminates the resolution degradation that was formerly caused by drifting in the linear biased amplifier.

C. Hardware development of a prototype system for transferring spectra from multidimensional analyzers to computer-readable magnetic tape has been completed. The transfer is verified by subtracting the spectrum on tape from the memory. This is a sensitive verification of the operation of each component in the system, because each of the 4096 channels is zero at the end of the verification step. Any nonzero value indicates a malfunction in one of the components. The controlling device for the transfer and verification system is a minicomputer. To obviate the need for adding peripherals to the controller for a short period of time to develop the controlling software, a cross-assembler was used on a computer with more extensive peripherals. The cross-assembler was developed by another ERDA contractor. A new driver program for a fast printer that is compatible with the cross assembler was developed, and it was used immediately to assemble programs for the spectrum transfer controller. Programs for the spectrum transfer system are being developed using structured programming techniques. The first set of programs allow transfer and verification from one of 15 analyzer memories. With minor changes to this basic program set, another set of programs will be added to give more data transferring capability such as moving a selected spectrum from the tape into the memory in an addition or subtraction mode to allow examination of spectral details. A third and final set of programs will allow identification parameters to be stored with the spectra.

Measurement of Ultra Trace Quantities
of Several Elements in CTR
Experiments

J. C. Langford

Various metals are being considered for use in construction of containment vessels for controlled thermonuclear reactors. Little is known about the rate of sputtering losses from the surfaces of metals due to high neutron fluences. This sputtering process may be a principal means of releasing radioisotopes from a CTR system to the environment. Because of the short half life of isotopes measured and the unusual chemistry at the collector surfaces, the following procedures were designed, tested and then used to give a satisfactory measure of the sputtered metal atoms present on the collector surfaces. Emphasis was on high chemical yields and rapid chemical separations.

Highly selective and efficient counting techniques were developed to obtain an accurate measurement of the sputtering rates from metal surfaces. High-purity metal discs were sandwiched between ultraclean collectors and irradiated at high neutron fluences. The collector discs were then submitted to surface leaching followed by rapid chemical separation and counting.

Europium, aluminum, chromium and molybdenum were all separated from the surface of silicon wafers by HF etching. A small quantity of perchloric and boric acids was added and

heated to volatilize BF_3 . Separations of Eu, Al, and Cr were made by $\text{Fe}(\text{OH})_3$ precipitation.

For Eu determination both Fe^{+++} and Al^{+++} carriers were added, the solution made basic to separate the soluble Al^{+++} complex, then the solution filtered through a 5 μ millipore filter under vacuum, and the filter then counted on Ge(Li) diode. The yield was $93\% \pm 5\%$.

For aluminum determination, Al^{+++} and Fe^{+++} carriers were added. The solution was neutralized with NH_4OH and filtered through a 5 μ millipore filter. The filter was counted on a 26% Ge(Li) diode. After decay of all interference, the Al on the filter was reactivated using Cf^{252} and counted. The method gave a yield of $81\% \pm 8\%$.

For Cr determination, Cr^{+++} and Fe^{+++} carriers were added, then Ba^{++} carrier and 30% H_2O_2 and the sample neutralized with care using NH_4OH . The sample was filtered through 0.45 μ millipore filter and the activity counted. The filtrate was saved and Cr^{+++} measured by atomic absorption to give a chemical yield of $81\% \pm 3\%$.

Molybdenum was leached using a concentrated nitric and hydrofluoric acid mixture. After boiling down to a small volume the solution was made ~ 0.5 N in H_2SO_4 and extracted in a 0.5% Benzoinoxine solution in CHCl_3 . After holding in the organic phase for 24 hr for ^{99}Tc buildup, counting gave an indicated yield of $84\% \pm 5\%$.

In Situ Monitoring

Systems for in situ analysis of radionuclides around storage areas and nuclear sites are being developed. The development and initial use of an intrinsic Ge system for the determination of Pu and Am in soils was completed. A method based on gamma-ray analysis of grazing animal thyroids was developed as a possible method for U and Th exploration using their ^{226}Ra and ^{228}Ra daughters.

In Situ Measurement of Plutonium and Americium in Soils

K. K. Nielson, N. A. Wogman and
R. L. Brodzinski

The detection of ^{239}Pu and ^{241}Am contamination on soil surfaces and in pond sediments can be accomplished by several methods including alpha counting or x-ray and gamma-ray counting. Some of the more promising methods were compared on the basis of sensitivity and suitability for in situ measurements.

Of the methods available, Alpha energy analysis with a $450\text{ mm}^2 \times 0.1\text{ mm}$ gold surface barrier detector provided the lowest detection limits, but required thin sample counting in an evacuated chamber. This method is not suitable for in situ field measurements in soils and sediments due to the short range of alpha particles in air and their much shorter range in soil.

Total ionizing radiation measured with a chamber having a 25 cm^2 window and containing a 90% argon, 10% methane gas mixture also provided low detection limits for ^{239}Pu and ^{241}Am sources. However, this method provides no information about the energy or type of radiation, and its in situ application is thus limited to locating contaminated areas. Identification of the nuclides and measurement of their activities and depths requires other methods.

Scintillation counting with a $1,000\text{-mm}^2$ surface area by 5-mm thick

NaI(Tl) crystal resulted in higher minimum detection limits than observed with other methods, but provided a means of identifying the presence of ^{241}Am through observation of its 59.5 keV gamma rays. While NaI(Tl) crystals are commonly used for gamma-ray analysis, they have insufficient resolution for identification and measurement of possible interfering photopeaks from natural radioactivity and fallout with its associated fission and activation products. They can thus be used for in situ ^{241}Am measurement only in the absence of interference.

The use of an intrinsic germanium diode for in situ x-ray and gamma-ray analysis offers positive identification of the radionuclides being analyzed as well as low detection limits. The detection limits measured with a $400\text{ mm}^2 \times 5\text{ mm}$ intrinsic germanium diode (Princeton Gamma Tech) are compared with those determined by the other methods in Table 1.6. The high resolution (400 eV at $\text{U L}\alpha$) of the intrinsic germanium diode permits easy identification of possible interfering nuclides as well as separate measurement of the four principal x-ray peaks.

Since the Ge detector offered the capability for specific measurement of ^{239}Pu and ^{241}Am , details of such measurements were further investigated. The effect of photopeak attenuation by the soil matrix was of primary concern, since activity slightly below the soil surface is determined less efficiently, giving

TABLE 1.6. Comparison of 3σ Detection Limits for 10 min Counts (dpm/cm²)

	²⁴¹ Am alone	²³⁹ Pu alone	²⁴¹ Am	²³⁹ Pu
			Combined: 0.078 Am/Pu activity ratio	
Total "beta" counts 25 cm ² chamber	6.1	4.4	not specific	
Alpha counts 450 mm ² x 0.1 mm detector	not measured	1.5 (vacuum) 13. (air)	not measured	
NaI(Tl) crystal 1000 mm ² x 5 mm	105.	266	122	694
Ge diode 400 mm ² x 5 mm	6.1	72	6.1	22

rise to possible erroneous interpretation. The magnitudes of these absorption errors for ²⁴¹Am measurements are indicated for various depths in Figure 1.8.

Since high-energy photopeaks suffer less absorption than low-energy ones, the ratio of ²⁴¹Am peak areas can be used both to estimate the average depth of the activity in the soil surface and to correct the activity measurement for attenuation by the soil. A number of soil, cellulose and aluminum oxide mixtures were prepared and pressed into wafers of varying thickness to determine the variation in peak ratio with the depth of the activity and with the average Z of the absorbing medium. The wafers were placed between the source (²⁴¹Am or ²³⁹Pu) and the detector for the attenuation measurements. The attenuation of the 26 keV x rays from ²⁴¹Am is plotted as a function of the peak area observed in Figure 1.9. One can thus use an intrinsic germanium diode for accurate surface soil activity and depth measurements of ²⁴¹Am and ²³⁹Pu by utilizing the ratios of ²⁴¹Am x-ray and gamma-ray peak areas. The variation in ²⁴¹Am detection limits with soil depth when the depth corrections are made is illustrated by the

FIGURE 1.8. ²⁴¹Am Detection Limits as a Function of Depth in the Soil Surface

solid line in Figure 1.8. The broken lines show detection limits based entirely on 60 keV peak measurements using the intrinsic germanium diode and the NaI(Tl) crystal.

FIGURE 1.9. Attenuation of 26 keV γ -Rays from ^{241}Am as a Function of the γ -Ray Peak Area Ratio (top), with the Effect of Varying Absorber Z on the Ratio at Various Depths (bottom)

Simultaneous measurement of ^{239}Pu and ^{241}Am can also be accomplished more precisely with the diode than by the other methods. Table 1.6 shows the detection limits for the combined measurements.

Recent advances in solid cryogen cooling methods would allow a commercially available "down-looking" intrinsic germanium detector to be used in field applications for periods over 24 hr without danger of detector damage due to warmup.¹ Electronics requirements (bias, amplification and pulse height analysis) for the diode detector are similar to those for a NaI(Tl) crystal.

Radium Accumulation in Animal Thyroid Glands...A Possible Method for Uranium and Thorium Prospecting N. A. Wogman and R. L. Brodzinski

The increasing uranium demand for nuclear energy generation requires the discovery of new uranium ore reserves on a worldwide scale. Biogeochemical and geobotanical prospecting are attractive techniques in areas of extreme ruggedness, dense vegetation or semidesert conditions where plants are deep rooted and can transfer

uranium and uranium progeny from depths of as much as 20 m to their leaves. Since the primary problems in geobotanical prospecting are sampling the various species and analysis of the low levels of radioactive materials, a technique which could provide areal integration of the botanical specimens and/or concentration of the activity from the uranium or its daughters, would be most helpful.

It has been reported that ^{226}Ra and ^{228}Ra , progeny of ^{238}U and ^{232}Th respectively, are concentrated in cattle thyroids.² Since cattle are herbivores, it is quite possible that a method of biogeochemical prospecting for uranium and thorium is possible based on the biological concentration of these radium isotopes in the thyroids of foraging cattle. This biogeobotanical survey method not only integrates a large amount of plant material but also concentrates the radionuclides of interest in the cattle thyroids.

The direct analysis of the radium isotopes has only recently been made possible through the development and utilization of highly sensitive gamma-ray spectrometer systems incorporating high efficiency Ge(Li) diodes surrounded by NaI(Tl) anticoincidence shields. Utilization of these systems permitted the determination of high ^{226}Ra and ^{228}Ra concentrations in 14 thyroids from unselected cattle slaughtered in Nigeria, West Africa. As listed in Table 1.7, thyroids collected during 1972, 1973, and 1974 contain ^{226}Ra concentrations of up to 59 dpm/g wet weight of thyroid tissue. Radium-228 concentrations were measured up to 29 dpm/g wet weight. These data are compared in Table 1.7 to selected U.S. and Columbia cattle thyroids containing on the order of 1 dpm/g. These radium concentrations are many orders of magnitude higher than that which would be expected from uranium in the thyroids since uranium in mammals is normally found at only the 0.01 to 0.02 ppm level.

These nondestructive analyses were performed to a precision of $\pm 15\%$ using 1,000 min counting periods on about 1 g samples. Since cattle thyroids typically weigh 20 g, the same data could have been obtained in 50 min counting periods using the

TABLE 1.7. ^{226}Ra and ^{228}Ra in Cattle Thyroids

Slaughter Date	Thyroid Sample (gm) (a)	d/m/gm		$^{228}\text{Ra}/^{226}\text{Ra}$
		^{226}Ra	^{228}Ra	
4/10/72 Nigeria	3.7	1.18	1.74	1.47
5/25/72 "	2.5	6.39	3.98	0.62
12/14/72 "	5.8	5.23	2.45	0.47
1/20/73 "	5.9	5.79	2.36	0.41
8/14/73 "	5.7	6.21	3.79	0.61
10/13/73 "	0.43	50.5	14.8	0.29
11/30/73 "	1.0	58.7	23.4	0.40
4/30/74 "	0.64	57.8	28.5	0.49
5/30/74 "	0.73	19.7	13.8	0.70
6/29/74 "	3.4	5.86	3.58	0.61
7/30/74 "	0.75	47.2	25.9	0.55
8/30/74 "	1.8	5.97	3.88	0.65
9/30/74 "	4.1	0.31	1.0	3.2
5/31/75 "	1.9	22.1	13.1	0.59
10/14/64 U. S.	-	1.18	1.04	0.88
4/28/65 U. S.		0.72	0.38	0.53
5/26/65 Columbia		0.90	1.35	1.5
5/26/65 Columbia		0.81	1.31	1.6
11/14/64 Columbia		1.35	1.80	1.3

a. The weights used for calculation were wet weights of trimmed tissues preserved with paraformaldehyde powder.

whole thyroid. In an exploratory survey, however, more than one thyroid would be used and sample sizes of 200 g or greater would allow analyses to be made at the same accuracy level in periods of 5 min or less. In this manner a rapid survey of the ^{226}Ra and ^{228}Ra concentrations in the thyroids of grazing animals

could be accomplished. Since these animals have known geographic ranges, a survey of the relative uranium and thorium concentrations in these areas could be immediately obtained. Since other animal thyroids should also demonstrate increased concentrations of radium, a variety of geographic ranges may be surveyed.

• ENVIRONMENTAL POLLUTION ANALYSIS

To understand the physical and chemical behavior of inorganic pollutants in the environment, it is essential that techniques be developed to sample and measure pollutants at their source, during their transport and throughout the environment. To meet some of these needs: 1) a method developed in 1974 was further refined for the in situ analysis of trace element pollutants in sediments by x-ray fluorescence for measurement of up to 30 elements at the 2-50 ppm levels; 2) plasma excitation techniques were advanced for chemical species determination of As and Sb; and 3) sampling methods and gas chromatograph techniques for inorganic and organic gases have undergone further development.

Analysis of Arsenic in Solution by X-Ray Fluorescence of Ferric Hydroxide Precipitate E. A. Crecelius

The As levels in natural waters usually range from less than one to several hundred ppb. The maximum allowable drinking water standard is 50 ppb. The analytical methods which are currently used for As measurements at these levels are time-consuming and expensive. Thus a reliable and relatively simple technique was needed to study the As concentrations and behavior in large numbers of natural water samples and to determine if abnormally high As levels are present.

A reliable, fast, inexpensive technique has been developed for the analysis of arsenic in natural water. The technique, which consists of coprecipitating As from solution with $\text{Fe}(\text{OH})_3$, then mounting the precipitate and quantifying the arsenic by x-ray fluorescence (XRF), has a detection limit of 3 ppb As in 50 ml of water. Briefly, the procedure consists of adding 4 mg of Fe^{+3} in dilute HCl to a 50 ml sample of filtered

water. The solution is adjusted to pH 6-8 using a few drops of 30% NH_4OH . The $\text{Fe}(\text{OH})_3$ precipitate is filtered onto a 25 mm diameter, 0.4 μ pore size membrane filter. After drying, the precipitate is analyzed for As by XRF. Care must be taken to correct for the PbL_{α} counts which interfere with the AsK_{α} line. Standards were prepared by spiking water samples which had already been precipitated and filtered. Blanks were prepared by precipitating $\text{Fe}(\text{OH})_3$ in water samples that had already been processed.

This technique, which requires approximately 10 min of mounting time and 10-30 min of counting time, can be applied to other elements such as antimony and selenium that are efficiently coprecipitated from water by $\text{Fe}(\text{OH})_3$.

An Improved Collimator for X-Ray Fluorescence Analysis K. K. Nielson and N. W. Wogman

A small diameter, tapered collimator has been developed to improve the capability to quantitatively analyze a small, non-uniform sample by

x-ray fluorescence in a region of uniform sensitivity. This new tapered collimator provides higher count rates and minimizes problems due to nonhomogeneity in larger samples.

Quantitative x-ray fluorescence analysis is often achieved by preparation of samples which are uniform over the area analyzed by the spectrometer. Particulate samples from air and liquids are thus collected by filtration while plant, dried or ashed tissue and soil samples can often be pressed into uniform thin wafers for reproducible analysis. Small quantities of liquid solutions present a preparation problem since they are not readily evaporated to uniform residues over areas of several square centimeters. Such samples must either use internal standards or be analyzed as a non-uniform residue enclosed in a region of uniform sensitivity in the x-ray spectrometer. The latter option is permitted by a simple modification to the x-ray spectrometer collimator which makes the instrument less sensitive to sample nonhomogeneity.

The instrument used in this study was a Kevex 810 tube excitation system which operates in the secondary excitation mode. Both the sample and the surrounding mounting were irradiated, but collimation of the Si(Li) detector (to 2.54 mm) allowed analysis of only the center region of the sample. Larger collimators were available, but their use allowed interference from the 35 mm slide frames used for sample mountings.

The uniformity and extent of the region being analyzed using various collimators was evaluated by analyzing a 1-mm cubic copper pellet in successive 1 mm positions along the major axes of the sample holder. The intensity of the Cu peak as a function of sample position was then plotted as equal-intensity contours as shown in Figure 1.10. The elliptical shape results from the 45° sample-detector geometry. The very small sample region over which the collimator offered uniform sensitivity (~ 3 -mm diameter for a 10% sensitivity drop) can be explained by the sample-detector geometry as illustrated in Figure 1.11. Region A on the sample plane is the region having a maximum solid angle of exposure to the detector. The intensity of the copper peaks when the

FIGURE 1.10. Intensity Map Comparison of Two Detector Collimators

FIGURE 1.11. Comparison of X-Ray Detector Collimator Geometries, Showing Regions of Uniform (A) and Non-uniform (B) Sensitivity

copper cube was in this region was thus nearly constant. Region B on the sample plane has a reduced solid angle of exposure to the detector due to the shadow of the top corner of the detector collimator. Copper peak intensities were lower when the sample was in this region.

The advantage to be gained using a longer, tapered collimator is also graphically illustrated in Figure 1.11. Additional length was included in the tapered collimator design to minimize the size of the B region, hence minimizing x-ray detection from the sample mounting.

The tapered collimator was made of iron, with an aluminum insert and base. The aluminum served to absorb any x ray from iron excited in the collimator. Analyses using the collimator with a wide variety of sample types and thicknesses indicate it does not contribute any significant peaks which could be considered part of a "system blank."

Analyses comparing the two collimators show higher count rates for the tapered collimator, despite its smaller diameter. All comparisons were thus normalized to equivalent count rates. Analyses of a 63 mg/cm² NBS orchard leaf pellet (SRM 1571) showed a slightly higher peak/background ratio for the high energy end of the x-ray spectrum using the tapered collimator. It is presumed that the improvement was not noted for low energy x rays due to the longer x-ray path length in the region of the sample most distant from the detector. This region, corresponding to the right side of the ellipses in Figure 1.10, can readily be eliminated by inserting a shield at the top of the collimator.

The primary advantage gained with the tapered collimator, however, is the capability to quantitatively analyze a small, non-uniform sample in a region of uniform sensitivity. Problems due to inhomogeneity in larger samples are also minimized since a larger quantity of the sample is analyzed.

Trace Element Analysis of Natural Waters Using Energy Dispersive X-Ray Fluorescence

K. K. Nielson

The rapid, multielement analysis capability of energy dispersive x-ray fluorescence methods has been applied to the analysis of natural water samples. Since the x-ray fluorescence method has been found to have detection limits of several nanograms per square centimeter for many elements in air filter samples, excellent detection limits were predicted for water samples if the elemental content of interest could be removed and prepared as thin, uniform deposits. Both thionalid and ammonium-l-pyrroline dithiocarbamate (APDC) have been found to give good recoveries of added trace elements from water by coprecipitation and precipitation, respectively.

The x-ray fluorescence apparatus used for these studies was a Kevex Model 810 tube excitation system operating with secondary excitation.

The use of thionalid has been described for the preparation of water samples for neutron activation analysis.³ The sample preparation methods used in our x-ray fluorescence work described here were similar to those reported, and involved: 1) addition of a dilute thionalid solution to the filtered sample, 2) heating the sample to boiling, 3) cooling, and 4) filtering. The method appeared to be equally useful for seawater and freshwater samples. The voluminous thionalid precipitate serves as a self-supporting sample, eliminating the need for a membrane sample backing. Nearly complete recovery was obtained for standard additions of 1-5 ppb ($\mu\text{g}/\text{l}$) Cr(VI) in sea water, with a 3 σ detection limit of 0.17 ppb. Contamination problems have not yet allowed measurements of Cu and Pb at the ppb concentration level.

The use of APDC to precipitate dissolved metals required minimal sample preparation time. Approximately 20 mg of APDC powder were added directly to a 1 l water sample at room temperature. After adjustment of the

pH to 2.8 and brief stirring (≈ 10 min), the sample was filtered through a 0.5μ Nucleopore membrane filter, and the filter was mounted for analysis. Standard additions of Ni (10-20 ppb) to freshwater samples were completely recovered by this method. The 3σ detection limits for Ni estimated from spectral backgrounds for APDC sample analyses was 0.15 ppb. This limit is based on 10 min analyses of 1 g samples filtered through a 9.6 cm^2 filter area. Excellent recovery was also achieved for Cu(II), while Cr(VI) and Pb(II) were incompletely recovered, possibly due to differing pH requirements.

The use of APDC has recently been reported in connection with x-ray fluorescence analysis of Fe, Cu, Zn, Hg and Pb in freshwater samples.⁴ While recovery ratios for Fe and Zn at a pH of 2.0 were low (0.59 and 0.21), those for Cu, Hg and Pb were near 1.0 for 100 ppb concentrations. Salinities typical of sea water were also reported to be compatible with APDC precipitations.⁴ Since more than 30 transition metals form insoluble precipitates with APDC,⁵ it appears that this method or variation of it may provide an extremely sensitive and simple method for multielement x-ray fluorescence analysis of water samples.

Corrections for X-Ray Absorption in Aerosol Samples Using X-Ray Scattering

K. K. Nielson

A method has been developed for estimation of self-absorption correction factors for x-ray fluorescence analysis of aerosol samples utilizing coherent and incoherent scatter peaks from the x-ray source. Since the correction method can be applied to existing data reduction programs, absorption corrections can be rapidly made. While secondary excitation was used for the analyses described here, the method should also apply to direct excitation by x-ray tubes or isotopic sources. Since the scatter peak data are collected simultaneously with energy dispersive x-ray spectra, the method requires no additional analyses to determine the correction factors.

Reliable methods for determining self-absorption correction factors have often depended on transmission

measurements for evaluating the sample mass absorption coefficient and additional analyses to determine an average depth of sample deposition in the filter.⁶ Multiple analyses using different excitation x-ray energies have also been used to estimate particle self-absorption effects.⁷ The methods are generally time consuming, however, since they require multiple analyses of each sample.

A simple relationship has been found to approximate the correction factor, F_i , utilizing the usual exponential function

$$F_i = \frac{\mu_i S}{1 - \exp(-\mu_i S)} \quad (1)$$

where μ_i represents the effective mass absorption coefficient for an x-ray of energy E_i (keV), and S is related to the sample thickness. Since plots of $\log \mu_i$ versus $\log E_i$ are linear with similar slopes in the energy and element range considered here, the μ_i are computed as

$$\mu_i = E_i^{(a)} \exp(b), \quad (2)$$

where a is the slope and b is the intercept of the $\log \mu_i$ versus $\log E_i$ plot. The value of b is chosen as that obtained for an absorbing matrix of oxygen. Since the value of b cannot be expected to be invariant over all samples, adjustment for variation in the absorbing matrix is made using the scatter peak-determined value of S . The value for S (gm/cm^2) is determined as

$$S = fC^2/It + g, \quad (3)$$

where C is the coherent scatter peak area, I is the incoherent scatter peak area, t is the analysis livetime (min), f is the slope of the thickness versus C^2/It line ($\text{gm}\cdot\text{min}/\text{cm}^2\cdot\text{counts}$) and g is the intercept (g/cm^2). The coefficients f and g are determined by analysis of samples whose absorption corrections are known.

A simple calibration and demonstration of the method is accomplished

using 2 μ particulate zinc sulfide samples collected on Whatman 41 and IPC filters. The large absorption difference between zinc and sulfur x rays permits accurate measurement of sulfur absorption corrections from the Zn:S ratio. Figure 1.12 illustrates the precision and accuracy of some sulfur absorption corrections thus obtained. Precisions for the sulfur corrections were in the range of 10-20% relative uncertainty. X-ray peaks of higher energy than sulfur would, of course, suffer much less error from uncertainty in their absorption corrections.

Although the correction method is an approximate one, it yields specific correction factors for each sample, thus facilitating the analysis of air filters having wide variations in particulate loading. Calibration of the constants f and g should be done specifically for a given type of filter on a given x-ray spectrometer, since the constants depend on the average depth of the particulate sample in the filter matrix. By adding the simple correction calculations to an existing data reduction program, absorption corrections are made with virtually no additional effort or time on the part of the analyst.

FIGURE 1.12. Estimated Self-Absorption Correction Factors for Sulfur Compared to Those Based on the Known Zinc-Sulfur Ratio

Application of Mercury Speciation Traps for Geothermal Gases E. A. Crecelius

One of the major environmental concerns of the geothermal power industry is the release of mercury to the environment. The concentration of Hg in geothermal gases on a water-free basis is known to exceed 1 mg/m³. While the major form of Hg in the noncondensable geothermal gases is elemental Hg vapor, other specific forms of Hg have not been determined. Since the bioavailability and toxicity of Hg compounds varies greatly with chemical form, it was essential that we develop and evaluate methods that can be used to determine the specific form of Hg in geothermal fluids.

Mercury speciation traps have been evaluated in the laboratory and used to characterize the different forms of Hg released by geothermal power plants. While other workers have developed a solid sorption trap for specific forms of Hg, they have not applied these techniques to the unique chemical environment of geothermal power. We have evaluated and modified some of these traps for use in geothermal gases. The traps used included: Chromosorb-W coated with SE-30 and HCl vapor (trap removes Hg⁺² compounds and passes methylmercury chloride, dimethylmercury and elemental mercury), Chromosorb-W coated with NaOH (trap removes methylmercury chloride and passes elemental mercury and dimethylmercury) silvered glass beads (trap removes elemental mercury), and gold-coated glass beads (trap removes dimethylmercury).

The four traps were connected in series and sampled a known volume of gas at ambient temperature. They were then heated during the analysis sequentially to specific temperatures from 200 to 500°C and Hg driven to a gold-coated glass bead transfer tube which in turn was heated to 500°C, driving the material to a helium plasma D.C. emission detector. The detection limit of the system is 1-ng Hg and the working range is 1-1,000 ng Hg. The possibility of compounds such as H₂S, NH₃ and water vapor interfering with the measurement of mercury and its compounds was examined and found to be unimportant at the levels normally observed in geothermal gases.

Analysis of Trace Elements in Sea
Water by Anodic Stripping Voltametry
E. A. Crecelius and J. C. Kutt

Anodic stripping Voltametry (ASV) is an electroanalytical technique that has been used for the direct determination of Cu, Pb and Cd in natural waters. The ASV method is analytically attractive for pollution studies involving environmental samples because the measurements are direct, rapid, and sensitive to ppb levels.

During the last year two types of electrodes have been evaluated for performance on seawater samples. A wax impregnated graphite electrode (WIGE) was used in a rotating cell containing sea water adjusted to a pH of 4.7 which was purged with CO₂ gas to remove O₂. Mercuric chloride solution was added to the sample and

the Hg and trace metals were plated on the electrode for 5 min then stripped into the solution. This technique detects less than 0.1 ppb of Cu, Pb or Cd in a 15 ml sample of sea water. A disadvantage of the WIGE is that at a pH of less than 2 the electrode rapidly deteriorates. The standard hanging mercury drop electrode was also evaluated. This electrode worked well at metal concentrations greater than 1 ppb and at pH ranges from 1-8.

This ASV technique is allowing the stability constants of metal-inorganic as well as metal-organic complexes in sea water to be determined. The quantity of metals complexed by organic matter is being determined by analyzing samples before and after the dissolved organic matter is destroyed by UV light oxidation.

• IN SITU POLLUTANT MEASUREMENTS

The program is for development and demonstration of instrumental technology for the in situ analysis of a wide spectrum of inorganic, organic and radionuclide species in oceans and freshwater sediments.

In Situ X-Ray Fluorescence Analyzer Improvements N. A. Wogman

The existing in situ x-ray fluorescence analyzer was redesigned by changing its specialized F.E.T.'s, preamp and high voltage supplies to allow resolution to be maintained in its underwater operation equal to that of a laboratory system. The present 80 mm² Si(Li) system resolution through 100 m of underwater cable is 195 eV at 5.9 keV. The bias is now preset without any adjustment being possible. The necessary power which must be delivered from the surface vessel to the underwater system is +30 volts and -24 volts. This power supply and F.E.T. modification has allowed the use of standard optical preamp electronics. This procedure increased the resolution and the reliability of the in situ system.

The present 195 eV resolution has allowed the sensitivity (Table 1.8) of the underwater system to approach within a factor of two that obtainable with laboratory x-ray fluorescence analysis. Figure 1.13 shows spectra obtained with the in situ system of freshwater and marine sediments from Lake Washington and Puget Sound in the State of Washington. As shown in Table 1.9, this instrument allowed the detection of 17 elements ranging from the tens of ppm to percent levels with a 360 sec count. The original excitation source used contained 18 of ¹⁰⁹Cd but has been replaced with a 55 mCi ¹⁰⁹Cd source. The increased count rate dead time due to the stronger source is acceptable with pulse pileup rejection operation allowing resolution to be maintained with this intense source.

TABLE 1.8. Detection Sensitivities and Prominent X-Ray Energy Lines for the In Situ Seabed Analyzer

ELEMENT	SENSITIVITY*		PROMINENT X-RAY ENERGIES**										
	(ppm)		(kev)										
		K α	K β										
Cr	2-5	5.412	5.947										
Mn		5.895	6.490										
Fe		6.400	7.058										
Co		6.925	7.649										
Ni		7.472	8.265										
Cu		8.041	8.905										
Zn		8.631	9.572										
Ga		9.243	10.262										
Ge		9.876	10.980										
As		10.532	11.723										
Se	4-10	11.209	12.493										
Br		11.909	13.288										
Rb		13.375	14.956										
Sr		14.143	15.830										
Y		14.933	16.732										
Zr		15.747	17.661										
Nb		16.584	18.614										
Ag		22.105	24.927										
Cd		23.110	26.078										
In			24.140	27.257									
		Li	L α 1	L β 1	L β 2	L γ 1	L γ 2	K α 2	K α 1	K β 1,3	K β 2		
Sn	4-10	3.045	3.444	3.663	3.905	4.131	4.377	25.196		28.465	29.109		
Sb	5-20	3.189	3.605	3.844	4.101	4.348	4.600	26.276		29.702	30.387		
Ba		3.854	4.466	4.828	5.156	5.531	5.797	31.817	32.194	36.341	37.257		
La		4.124	4.651	5.042	5.384	5.788	6.060	33.034	33.442	37.761	38.730		
Ce		4.287	4.840	5.262	5.613	6.052	6.325	34.279	34.720	39.214	40.233		
Au	2-5	8.494	9.713	11.442	11.585	13.382	13.709						
Hg		8.721	9.989	11.823	11.924	13.830	14.162						
Pb		9.184	10.551	12.614	12.623	14.764	15.101						

* BASED ON 3 σ DETECTION LIMITS USING A 5 MINUTE ANALYSIS AN 80 mm² Si (Li) DIODE AND A 100 mc¹⁰⁹Cd SOURCE

** APPROXIMATE RELATIVE INTENSITY RATIOS WITHIN SERIES OF A GIVEN ELEMENT ARE (K α 1=100):

K α 1 = 100	L α 1 = 100	L β 4 = 3-5	M α 1,2 = 100
K α 2 = 50	L α 2 = 10	L γ 1 = 1-10	M β = ~60
K α 1,2 = 150	L β 1 = 50	L γ 3 = 0.5-2	
K β 1 = 15-30	L β 2 = 20	Li = 1-3	
K β 2 = 1-10	L β 3 = 1-6	LN = 1	
K β 3 = 6-15

FIGURE 1.13. In Situ X-Ray Fluorescence Spectra of Freshwater and Marine Sediments

TABLE 1.9. Elemental Concentrations of Selected Marine and Fresh Water Sediments Analyzed by the In Situ X-Ray Fluorescence Analyzer

Element	Marine (ppm)	Fresh Water (ppm)
Ti	0.49%	0.3%
Cr	450	
Mn	820	
Fe	6.9%	5.2%
Ni	100	
Cu	200	250
Zn	315	0.1%
As	18	
Br	150	30
Rb	120	30
Sr	245	350
Y	53	220
Zr	215	190
Nb	27	15
Hg	<9	<10
Pb	175	5500

An Improved Method for Automatic Peak Analysis in X-Ray Spectra
K. K. Nielson

An automated data reduction system employing a two-point integration technique known as the width-determined background (WDB) method provides the reliability and convenience to analyze large numbers of samples economically using x-ray fluorescence.

A problem of concern in automating energy dispersive x-ray spectral analysis procedures is that of obtaining reliable photopeak areas from the digital data collected from analyses of environmental samples. A variety of peak analysis methods for gamma-ray spectra, which have been adapted for use in x-ray spectral analysis, have generally involved background subtraction followed by peak location and peak shape fitting. This approach often fails with x-ray spectra due to inaccurate background estimation. X-ray spectra having many peaks frequently have insufficient data between peaks to determine the background magnitude; thus the gamma-ray programs, although reliable on large or isolated x-ray peaks, fail in measuring the areas of significant small peaks. In trace element analysis for environmental baseline studies, such peaks are vitally important. Since modern Si(Li) x-ray spectrometers do not suffer from the gain shifts common to gamma-ray spectrometry, precise measurement of peak position is also unnecessary.

A method, called the width-determined background (WDB) method and based on the familiar two-point integration technique, is illustrated in Figure 1.14. The WDB method relies on measuring only the top part of the peak, as defined by the intersection of a peak-centered, fixed-width window with the data points. The area (A) can be shown to be background independent, and relies only on constant detector resolution. A simple linear variation of the window width (W) with energy adequately allows for width variations. Individual peak shape and width differences (i.e., K versus L x-rays) are accounted for in the calibration as long as the same width is always used to analyze a given peak.

The WDB method is reliable even for small peaks when several normally variable peak parameters are held fixed. X-ray peak location can be precisely determined by previous energy calibration, and need not be allowed to vary. Peak width is reproducible, allowing direct analysis of a given element. The background parameter may also be considered fixed since it depends entirely on the fixed width parameter and the given data. One thus avoids depending on several channels at the sides of a peak to estimate the true background.

Since the WDB method utilizes a smaller fraction of the peak area for measurement, it has statistically

FIGURE 1.14. Comparison of Peak (A) and Background (B) Regions as Defined in Two Peak Analysis Methods

poorer precision than the background subtracted methods. Figure 1.15 evaluates the precision of the two methods as a function of widths for a small peak with a height/background ratio of 1.0. A width of approximately 3.4σ has been found to minimize effects of overlapping peaks while giving adequate precision. Since background is not dependent on additional background-defining points, the accuracy of the WDB peak areas is greater than that routinely achieved with simple background-subtracted methods.

In practice, 26 peak regions in a 1024 channel spectrum are analyzed and interference-corrected elemental concentrations are computed in less than 1 min. Least-squares polynomial smoothing⁸ is used on square-root transformed data⁹ to allow precise interpolation of the intersection of the data with the analysis window. Surveillance of data reduction by the analyst is thus reduced to occasional checks on energy calibration.

FIGURE 1.15. Comparison of the Precisions of Two Methods of Peak Area Measurement as a Function of the Energy Window Width

• TRACE CONSTITUENT ANALYSIS BY LASER EXCITATION

A laser-induced fluorescence technique has been theoretically evaluated for the detection of low levels of ^{85}Kr with detection limits of less than 10^4 atoms. The scheme involves a two-photon Doppler-cancelled excitation of ^{85}Kr by laser radiation to a high energy electronic state. The atoms are then detected by the fluorescence emitted as the highly excited state decays to lower excited states.

The Analysis of ^{85}Kr by Laser Excitation Techniques

B. A. Bushaw and T. J. Whitaker

The onerous task of measuring radioactive and stable trace pollutants at low levels may be simplified by the use of laser excitation techniques which offer the possibility of severalfold increases in sensitivity. The application of this technique to the analysis of ^{85}Kr has been studied in the first stage of this program. By considering state energies, line shapes and transition probabilities, we are able to predict that better than one emitted photon may be detected for each 10^4 atoms of ^{85}Kr in the path of the exciting laser radiation. This report summarizes the results of the theoretical calculations. The technique involves the excitation of an atom of ^{85}Kr in its ground state to an upper excited state. In Figure 1.16 the electronic energy level diagram for Kr denotes the excitation from $1S_0$ ground state to the $1D_2$ excited state. Fluorescent decay then occurs to the $3P_2$ state. The wavelength of the photon emitted during this decay is approximately 0.76μ , which is detectable using conventional photon counting techniques.

Inspection of Figure 1.16 reveals that the excited states of Kr are in-

FIGURE 1.16. Electronic State Diagram for Kr Showing the Lower Lying States

accessible to single-photon excitations using conventional laser sources. However, multiphoton processes observe different selection rules than single-photon processes. For a two-photon absorption, the change in the electronic orbital

angular momentum and the total angular momentum¹⁰ (orbital + spin in L-S coupling) is equal to 0 or ± 2 . Figure 1.16 shows three states, 1S_0 , 3D_2 , 1D_2 , which satisfy these selection rules for excitation from the 1S_0 ground state. Due to experimental limitations, the 1S_0 is not accessible. Because more experimental information is available on the 1D_2 state, we have chosen this state for initial studies.

Two-photon transition probabilities are significantly lower than those for a single-photon process. Comparatively little is known about two-photon transition probabilities; however, the second order perturbation equation has been numerically evaluated for a similar system by Hansch, et al:¹¹

$$\Gamma = 7 \times 10^{-4} I^2 / \omega \quad (1)$$

where Γ is the two-photon transition probability, I is the intensity of the exciting radiation in watts/cm² and ω is the bandwidth of the laser in megahertz. For a laser bandwidth of 100 megahertz, a pulse duration of 500 nsec, and peak power of 3×10^5 watts/cm², the fraction of atoms excited in a single pulse, as given by Eq. 1, is 14%. The atoms excited to the 1D_2 state may then decay via a number of paths. From the measured lifetime of the 1D_2 state,¹² and the measured probability of the $^3P_2 \leftarrow ^1D_2$ transition,¹³ we find the predominant path to be radiation decay to the 3P_2 state. The percentage of excited atoms falling to this state is calculated to be 49%. With a conservative estimate of detection efficiency as 0.2%, the overall detection limit is calculated to be 1 in 7×10^3 .

Selective excitation of ^{85}Kr is an important aspect of this detection scheme. The bandwidths of the exciting radiation must be narrow compared to the separation between transition energies of different isotopic species. It is also necessary that the absorptions by different isotopes do not overlap. The results of a theoretical analysis of isotopic shifts and hyperfine structure of the 1D_2 state for ^{85}Kr and naturally occurring Kr isotopes is shown in Figure 1.17. Hyperfine splitting constants were determined from ex-

FIGURE 1.17. Hyperfine Structure of the 1D_2 State of Kr Calculated for ^{85}Kr and All Naturally Occurring Isotopes

perimental data presented by Rasmussen and Middleboe¹⁴ and theoretical work by McMillan and Halpern.¹⁵ Doppler widths have been calculated which are broader than the separation of ^{85}Kr and ^{83}Kr transition energies. Fortunately, it is possible to reduce Doppler broadening to a negligible amount by introducing two counterpropagating laser beams of different frequencies which will be simultaneously absorbed by the sample.¹⁶ The frequencies, ν_1 and ν_2 , will be such that $\nu_1 + \nu_2$ is the frequency of the transition, but neither $2\nu_1$ nor $2\nu_2$ coincides with the absorptions of any of the isotopes.

Figure 1.18 shows a schematic diagram of a system which will produce the required exciting radiation. The laser oscillator cavity tuned with a diffraction grating (DG) and three Fabry-Perot etalons (FP) will produce the frequencies $1/2\nu_1$ and $1/2\nu_2$. These may be separated using a polarized prism (P) and a quarter wave plate ($\gamma/4$) in conjunction with another Fabry-Perot etalon. Once separated the two frequencies are amplified

FIGURE 1.18. Experimental Arrangement for Selective Laser Excitation and Fluorescence Detection of ^{85}Kr

and frequency doubled with potassium pentaborate nonlinear crystals (SHG) to produce ν_1 and ν_2 . After filtering (F_d) to remove the fundamentals ($1/2\nu_1$, and $1/2\nu_2$), ν_1 and ν_2 are directed at the sample from opposite directions. The resulting fluorescence signal from the sample is isolated with a line filter (F_1) and detected with a photomultiplier.

• ANALYTICAL TECHNIQUES FOR MEASUREMENT OF ^{99}Tc IN ENVIRONMENTAL SAMPLES

A mass spectrometric technique is being developed for measurement of ^{99}Tc in environmental samples which promises to be 100 to 1000 times more sensitive than other methods. The feasibility of high sensitivity measurements has been demonstrated by the successful detection of as little as 10^{-14} grams of ^{99}Tc . The basic chemical separation and purification steps are being worked out for both water and vegetation samples. Sub-picogram measurements of electrodeposited ^{99}Tc have also been made.

Ultrasensitive Measurement of ^{99}Tc by Mass Spectrometry J. H. Kaye

From an environmental viewpoint ^{99}Tc is one of the most troublesome radionuclides produced through nuclear fission. This is due to the long half-life (2.13×10^5 years), the volatile nature of the heptoxide, the mobility of the pertechnetate ion in aqueous solution, the high fission yield (over 6%) and the affinity of several organs of the human body for this element. Furthermore, technetium is almost impossible to detect at low levels by direct counting methods since it emits only low energy beta radiation. The purpose of this study is to develop, by means of mass spectrometry, much more sensitive methods for its detection than are presently available. Currently planned ecological studies within existing DBER programs are awaiting development of the ^{99}Tc analytical procedures.

Highly sensitive measurement of ^{99}Tc was found to be possible by utilization of mass spectrometric techniques. The greatest sensitivity was achieved by placement of the ^{99}Tc directly onto the ionizing filament of the mass spectrometer and measuring the signal due to free metal ions

($^{99}\text{Tc}^+$). Quantities as small as 10 femtograms (10^{-14} g) of technetium as aqueous ammonium pertechnetate solution have been successfully detected in this manner. This amount is 1000 times less than can be detected by thermal neutron activation analysis and thus clearly demonstrates the potential measurement sensitivity of this approach.

In order to make a quantitative mass spectrometric measurement of technetium, it must first be chemically separated, highly purified and mounted onto a very small mass spectrometer filament. We have worked out and tested separation schemes for use with water and vegetation samples which result in 50-75% of the ^{95}mTc tracer added to the sample being recovered and loaded onto a rhenium filament. The initial steps for water samples involve either direct loading onto anion exchange resin or co-precipitation with copper sulfide followed by solvent extraction. For vegetation, the samples are ashed at low temperature, then fused with sodium carbonate and the melt dissolved in dilute acid. Further purification for both sample types includes precipitation of hydroxides, anion exchange, cation exchange, solvent extraction, and finally electrodeposition of technetium as the dioxide.

Major interferences in the mass spectrometric measurement have been observed for only two elements: molybdenum and ruthenium. Ruthenium interferes directly with ^{99}Tc since it has an isotope of mass 99 with a 12.7% abundance. Molybdenum does not interfere at mass 99, but its isotopes of masses 97 and 98 complicate the use of either ^{97}Tc or ^{98}Tc as isotopic tracers. Tests employing tracers and atomic absorption analysis methods indicate that the solvent extraction step results in a decontamination of more than 1000-fold for both ruthenium and molybdenum, and the anion exchange step results in a decontamination of 1000-fold for molybdenum and more than 100-fold for ruthenium. The hydroxide scavenge step is also highly effective for removal of ruthenium.

Techniques have been developed for electrodeposition of technetium as TcO_2 onto the very tiny cathode surface (5 mm^2) of a rhenium filament. Yields of over 95% are obtained in a few hours' plating time. After plating, the filament is heated in hydrogen to reduce TcO_2 to the less volatile technetium metal. This also removes traces of organic compounds and other volatile substances which may be present in small amounts. Mass peaks due to ruthenium were observed when a platinum anode was employed for electrodeposition but this was essentially eliminated by

substitution of a graphite anode. Molybdenum contamination has been observed from the electrodeposition step despite the exclusive use of teflon and other nonmetal materials for construction of the plating cell. However, the observed ion signal intensity appears to reflect more the tremendous sensitivity of thermal ionization mass spectrometry for molybdenum than the fact that a large amount of molybdenum is actually present. Even the best available sources of de-ionized or triply distilled water cannot be used for electrodeposition without special purification because the molybdenum contamination levels are otherwise higher by at least a factor of 100.

Work to date on this project has shown the feasibility of establishing a highly sensitive analytical procedure for ^{99}Tc . It has also shown what further work remains to be done on the chemical procedures and mass spectrometric techniques in order to develop a proven, integrated procedure. This work includes establishment of blank levels at masses 97 and 99, reduction of molybdenum interference, development of the isotopic dilution technique employing ^{97}Tc tracer, improvements to the mass spectrometer scanning and data output system and final testing of a working procedure by analyses of field samples.

• FAST REAL-TIME MEASUREMENT OF EXTREMELY LOW-LEVEL AIRBORNE PLUTONIUM

A mass spectrometer is being developed for monitoring low-level airborne plutonium on a real-time basis. The instrument will be capable of measuring plutonium concentrations below the MPC level. A direct air-inlet and a detector/readout system for the spectrometer have been developed. Design of the instrument has been completed and construction is in progress.

Fast Real-Time Measurement of Low-Level Airborne Plutonium

J. J. Stoffels and C. R. Lagergren

Current techniques for monitoring airborne plutonium are limited by the rate of radioactive decay which, for ^{239}Pu , amounts to only one disintegration per sec for every 10^{12} atoms present. A new technique using a direct-inlet mass spectrometer is being developed which will measure one ion count for approximately every 10^2 atoms of plutonium present and do this on an individual particle basis. A direct air-sampling inlet with a capacity of 5 standard cubic centimeters of air per sec has been developed for the spectrometer. A device to integrate and readout the ion current has been designed, constructed and tested. Design of the entire instrument is complete and construction is proceeding on schedule.

The direct-inlet mass spectrometer (DIMS) will be a desk-sized instrument (see Figure 1.19). Particulate material in the stream of air sampled through the capillary inlet will strike a hot filament in the ion source and produce a burst of ions lasting a fraction of a second. The ions will be mass analyzed in the magnetic field of the spectrometer and an ion detector will measure ions at mass 271, corresponding to $^{239}\text{Pu}^{16}\text{O}_2$.

The direct-inlet we have developed is shown schematically in Figure 1.20. It consists of four elements:

1. A capillary nozzle through which the ambient air is drawn generating a particle beam with small divergence;
2. A "skimmer" which skims off the bulk of the air expanding into a vacuum of 1 Torr while simultaneously transmitting the particle beam;
3. A collimator through which the particle beam also passes and by which another stage of differential pumping is accomplished; and
4. A hot filament to ionize the particles.

This inlet design evolved from initial work on an arrangement which did not include the collimator. Although simpler, that arrangement required 10 times more pumping capacity in the ion source chamber for the same sampling capacity.

An ion detector having a dynamic range in current measurement capability of approximately 10 decades has been developed for the Pu DIMS. This dynamic range will permit measurement of particles over a 3-decade

FIGURE 1.19. Direct-Inlet Mass Spectrometer

FIGURE 1.20. Direct Air-Sampling Inlet for a Surface Ionization Mass Spectrometer

range in physical diameter. The detector (see Figure 1.21) is basically of a type in daily use on PNL laboratory mass spectrometers. It converts an ion to a burst of secondary electrons which in turn produce light in a plastic scintillator. The light is

sensed by a photomultiplier which delivers an output pulse for each ion.

In addition to the normal pulse-counting mode of operation, which is useful for ion currents of 10^{-20} to 10^{-13} ampere, the detector has been

modified so that the aluminum coating on the scintillator can be used as a Faraday collector of the secondary electron current. This DC current can be measured with a conventional electrometer extending the range of ion current measurement to about 10^{-9} ampere.

This modified detector was tested and found to be capable of operation over the desired dynamic range. It gave a linear response for the two modes of operation which overlap in measurable ion current by more than one decade.

A prototype device has been designed and constructed to integrate the detector DC current from an ion-burst event and to read out the resultant data. The integrator has been satisfactorily tested with a rapidly-switched ion beam to simulate ion bursts. Plans have also been formulated for a minicomputer-based data-logging and readout system to handle the mass spectrometer analytical data.

An aerosol generator has been constructed to produce particulate aerosols of various materials. The aerosol generator has been used in the development of the inlet and will also be used for testing the entire spectrometer, when completed.

The detailed design of all components for the Pu DIMS is complete. Most components have already been fabricated. All commercially available equipment for the instrument has

FIGURE 1.21. Ion Detector with Simultaneous Pulse Counting and DC Current Modes of Operation for Wide Dynamic Range Response

been received or ordered. Completion of the instrument is anticipated during the third quarter of FY 1976. An intensive program of performance testing and calibration will then begin.

• ENVIRONMENTAL APPLICATION OF HOLOGRAPHY

This is a new project, supported by EPA pass-through funds received in June of 1975. Holography has the unique capability of photographically capturing the three-dimensional distribution of a scene for study at leisure. This project is aimed at evaluating this capability for the study of aerosols; their size, distribution and velocity. Detailed information of this type will be useful for studying the behavior of pollutants as they disperse, settle or re-entrain into the atmosphere. It should also be useful for inhalation toxicology, where knowledge of the aerosol generation and inhalation pattern is important.

Holography using short laser pulses is being studied to determine resolution capability and to determine optical arrangements most suitable for this application. A Korad 1500 ruby laser without Q-switching, is being used for initial studies. Q-switching and frequency doubling will be added later to study fast-moving aerosols.

Application of Holography to Environmental Studies B. P. Hildebrand

Holography, invented in 1948, is a photographic technique capable of recording a scene in three dimensions. This record, the hologram, can be used to reconstruct the scene for later study. The reconstruction is so perfect that it can be used to make precise measurements by means of interferometry, for example. The invention of the laser in the 1960's resulted in a rapid expansion in research of holography and the search for applications. The pulsed laser, in particular, imparted a momentum in the direction of particle holography since transient events could be recorded and studied later in the laboratory.

OBSERVATIONS

A Korad 1500 giant pulse laser is being used to determine the resolving

capability and optimum optical arrangements for aerosol investigations. At this stage of the investigation, the laser is not Q-switched resulting in an optical pulse length of about 0.5 msec. The wavelength of the radiation is 0.6934μ , resulting in a theoretical resolving capability of about 0.69μ . Actual resolution is less than this due to imperfections in the optical components and laser coherence.

A three-bar resolution chart was used to determine the best optical arrangement for aerosol investigations. These studies have shown that the in-line arrangement shown in Figure 1.22 is optimum. The smallest bar spacing in the target is 2μ and we have been able to resolve these. The hologram image is displayed as shown in Figure 1.23. The image may be examined with a microscope, or cast onto a television camera target for display on a monitor. The latter

FIGURE 1.22. Holographic System for Recording the Aerosol Cloud

system offers both optical and electronic magnification of the image for ease of observation and measurement.

A simulation of an aerosol, paint sprayed on glass slides, yielded good images which could be used to measure particle size and shape. Three-dimensional distributions were simulated by stacking a number of such slides.

CONCLUSIONS

Pulsed holography holds the potential for detailed analysis of dynamic aerosols. Particle sizes down to one wavelength appear to be resolvable thus allowing a moving aerosol cloud to be analyzed with respect to size, shape, velocity, distribution and density.

FIGURE 1.23. Display System

• CHEMICAL REACTION KINETICS

Thermodynamic data has been collected for a suite of compounds and elements which are to be included in a practical model of the dominant homogeneous and heterogeneous reactions following combustion in coal-fired generator facilities. An experimental technique is described for the measurement and identification of the volatilizable components of fly ash.

Measurement of Combustion Products by Chemical Reaction Kinetics

J. A. Campbell, W. D. Felix and
N. A. Wogman

With the planned increase in coal-fired electric generating facilities, knowledge of the combustion related release and disposition of the more toxic volatile elements and compounds has become increasingly important. It is the goal of this new program to model the relationships which control the interaction of volatile components, the formation rates of the particulates and the reactions at the surfaces of particulates. The physical and chemical reactions of the coal combustion products are being studied from a point just outside the firebox to a point outside the flue stack where atmospheric dispersion predominates. Input parameters to the model include initial feedstock composition, flue gas temperature as a function of time and distance from the firebox, pressure, and velocity. The study initially addresses combustion products and their reactions.

These products are developed through the almost instantaneous oxidation in the firebox of the carbon and hydrogen from the finely pulverized coal. Many of the other coal elements are volatilized leaving a molten or ash particle which can then serve as a center for the ensuing condensation of the volatilized constituents. Toxic trace elements such as

mercury, arsenic, antimony, selenium, copper and zinc may assume their thermodynamically stable composition for the temperature and oxidizing conditions which exist. However, the reaction rates for the various competing products will determine the forms of these and other constituents in the vapor phase.

A computer program, based on the minimization of free energy is currently being used to determine the thermodynamically stable forms of gaseous and solid combustion products at given temperatures. The algorithm used is based on the fundamental condition that a mixture of gases and condensed constituents at instant temperature is in equilibrium when its free energy is minimized under mass balance constraints. The total free energy of a system composed of m gaseous species (assumed to be ideal) and $n-m$ pure condensed species having an activity coefficient of one is

$$F(\bar{y}) = \sum_{i=1}^m y_i \left[\left(\frac{F_{T^0}}{RT} \right)_i + \ln P + \ln \frac{y_i}{\bar{y}} \right] + \sum_{i=m+1}^n y_i \left(\frac{F_{T^0}}{RT} \right)_i$$

where y_i is the number of gram-moles

of species i and $\bar{y}$ is the total gram-moles of gaseous species, $\bar{y} = \sum_{i=1}^m y_i$.

The $\frac{\bar{R}T}{F^0}$ terms represent the free energy function of the pure components while the logarithmic terms represent the free energy of mixing for the ideal gases. The problem becomes one of finding the proper combination of the gas and solid components, y_i 's, at a fixed temperature and pressure so that $F(\bar{y})$ is minimized under the constraint that the number of gram-atoms, c_j , of each of the d elements in the system is specified, i.e.,

$$c_j = \sum_{i=1}^m a_{ij} y_i + \sum_{i=m+1}^n a_{ij} y_i$$

$$j = 1, 2, \dots, d$$

the coefficients a_{ij} represent the number of gram-atoms of the y th element in species. The computational procedure uses Newton's optimization method and satisfies the given constraints of the Lagrangian multipliers.

An illustration indicating one result of the program is shown in Figures 1.24 and 1.25. A coal consisting

of C - 69.5%, H - 4.82%, N - 1.17%, and S - 4.75% (weight percent) was burned with an $\frac{\text{oxygen}}{\text{fuel}}$ weight ratio of 2:1 (Figure 1.24) and 3:1 (Figure 1.25) from 300-1500°K, the typical temperature range in the stack at a constant pressure of one atmosphere. Fifteen gaseous products were examined, but the results are shown only for the major species.

As seen in Figures 1.24 and 1.25, different oxygen/fuel ratios produce different major species at equilibrium. The effect of the quantity of O_2 overshadows the effects due to temperature changes.

Although only gaseous products have been considered to date, the program has the capacity to determine the concentrations of condensed species and gaseous products from the coal elements. Since most of the reactions occurring in the firebox are not at equilibrium, a time variable will be introduced into the program so that concentrations can be calculated as a function of time at a constant temperature and pressure.

In order to substantiate results obtained from the above theoretical study, a furnace has been constructed

FIGURE 1.24. Major Products for Oxygen/Fuel 2:1 Wt Ratio

FIGURE 1.25. Major Products for Oxygen/Fuel 3:1 Wt Ratio

to study the behavior of toxic elements through the use of trace element analytical techniques. Neutron irradiated fly ash will be placed in an alumina crucible in the furnace shown in Figure 1.26 and heated to the desired temperature. The volatile radioactive species will condense on the glass beads which can then be subsequently analyzed. This will allow the measurement of reverse release rates for approximately 30 elements known to be associated with coal combustion.

In a similar fashion, the fly ash will be heated by the furnace and the volatile species directed into a quadrupole mass spectrometer which permits the complete characterization of the decomposition products.

FIGURE 1.26. Furnace for Heating Irradiated Fly Ash

REFERENCES

1. N. A. Wogman, H. G. Rieck and J. R. Kosorok, "In Situ Analysis of Sedimentary Pollutants by X-Ray Fluorescence," Nucl. Inst. Meth., 1975, vol. 128, p. 561-568, BNWL-SA-5204, 1974.
2. L. Van Middlesworth and W. L. Robison, "Thyroid Concentration of Barium and Radium," Int. J. Nucl. Medicine and Biology, vol. 2, pp. 1-4, 1975.
3. D. E. Robertson and R. Carpenter, "Neutron Activation Techniques for the Measurements of Trace Metals in Environmental Samples," NAS-NS 3114, 1974.
4. J. F. Elder, S. K. Perry and F. P. Brady, Envir. Sci. Tech., vol. 9, p. 1039, 1975.
5. H. Malissa and E. Schoffmann, Microchim. Acta., vol. 1, p. 187, 1955.
6. F. C. Adams and R. E. Van Grieken, "Absorption Corrections for X-Ray Fluorescence Analysis of Aerosol Loaded Filters," Anal. Chem., vol. 47, p. 1967, 1975.
7. R. D. Giaque, R. B. Garrett, L. Y. Goda, J. M. Jaklevic and D. F. Malone, "Application of a Low Energy X-Ray Spectrometer to Analyses of Suspended Air Particulate Matter," in Advances in X-Ray Analysis, vol. 19 (in press), LBL-4402.
8. A. Savitzky and M. J. E. Golay, "Smoothing and Differentiation of Data by Simplified Least Squares Procedures," Anal. Chem., vol. 36, p. 1627, 1964.
9. T. Inouye, "Square Root Transformation for the Analysis of Quantum Fluctuations in Spectrum Data," Nucl. Inst. Meth., vol. 91, p. 581, 1971.
10. D. A. Varshalovich, "Selection Rules for Multiquantum Transitions," Opt. & Spectros (Engl) vol. 25, p. 85, 1968.
11. T. W. Hänsch, S. A. Lee, R. Wallenstein, and C. Wieman, "Doppler-Free Two-Photon Spectroscopy of Hydrogen 15-25*," Phys. Rev. Lett. vol. 34, #6, p. 307, 1975.
12. D. A. Landman and R. Dobrin, "Lifetimes and Alignment Depolarization Cross Sections of the $(4p^{55p})^1D_2$ and 3D_3 Levels in Krypton," Phys. Rev. A, vol. 8, #14, p. 1868, 1973.
13. Anne Pery-Thorne and J. E. Chamberlain, "Transition Probabilities in the Spectra of Neutral Neon, Argon and Krypton," Proc. Phys. Soc. (London) vol. 82, p. 133, 1963.
14. E. Rasmussen and V. Middleboe, "Hyperfeinstruktur und Kernmomente von Kr^{85} ," Z. Physik, vol. 141, p. 160, 1955.
15. J. A. McMillan and T. Halpern, Hyperfine Interactions: Tables of Isotropic and Anisotropic Parameters for the Atoms Hydrogen to Bismuth, Argonne National Laboratory, ANL-7784, 1971.
16. B. Cagnac, G. Grynberg and F. Biraben, "Spectroscopie D'Absorption Multiphotonique Sans Effect Doppler," J. de Physique, (Paris), vol. 34, p. 845, 1973.

PUBLICATIONS AND PRESENTATIONS

Publications

Crecelius, E. A., "Analysis of Total Iodine in Milk by X-Ray Fluorescence and Iodide Electrode," Anal. Chem., vol. 47, pp. 2034-2035, 1975.

Crecelius, E. A., "The Geochemical Cycle of Arsenic in Lake Washington and Its Relation to Other Elements," Limnol. Oceanogr., vol. 20, pp. 441-451, 1975.

Crecelius, E. A., M. H. Bothner and R. Carpenter, "The Geochemistries of Arsenic, Antimony, Mercury and Related Elements in Sediments of Puget Sound, Washington," Environ. Sci. Tech., vol. 9, pp. 325-333, 1975.

Mangelson, N. F., G. M. Allison, J. J. Christensen, D. J. Eatough, M. W. Hill, R. M. Izatt and K. K. Nielson, "Rapid Multi-Element Analysis of Biological Samples Using an Energy Dispersive X-Ray Fluorescence Method," in Trace Element Metabolism in Animals - II, Ed., W. G. Hoekstra, J. W. Suttie, H. E. Ganther and W. Mertz, Univ. Park Press, pp. 439-441, 1974.

Mangelson, N. F., K. K. Nielson, M. W. Hill, D. J. Eatough and L. D. Hansen, "A Potpourri of Proton Induced X-Ray Analyses," Third Conference on Application of Small Accelerators, Ed., J. L. Duggan and I. L. Morgan, North Texas State Univ., 1974.

Nielson, K. K., M. W. Hill, and N. F. Mangelson, "Calibration and Correction Methods for Quantitative Proton-Induced X-Ray Emission Analysis of Autopsy Tissues," Advances in X-Ray Analysis, vol. 19 (in press).

Rancitelli, L. A., R. W. Perkins, W. D. Felix, and N. A. Wogman, "Solar Flare and Lunar Surface Process Characterization at the Apollo 17 Site," Proc. of the Fifth Lunar Conference, Geochim. Cosmochim. Acta, vol. 2, pp. 2185-2203, 1974.

Wogman, N. A., "Comparison of Ge(Li) and Anti-Compton Systems for Measurements of Environmental Samples," IEEE Transactions on Nuclear Science, 1973 Symposium, vol. NS-21, no. 1, BNWL-SA-4876, February 1974.

Presentations

Carpenter, R. and E. A. Crecelius, "Geochemical Cycles and Budgets of Arsenic in Puget Sound and Lake Washington," NSF-RANN Trace Contaminants Abstracts, 2nd. Annual Conference Issue, August 1974.

Cooper, J. A., H. L. Nielson, N. A. Wogman, and R. W. Perkins, "Feasibility Study of In Situ Sediment Analysis by X-Ray Fluorescence," Nucl. Tech., vol. 26, pp. 224-231, June 1975.

Crecelius, E. A., "The Geochemical Cycle of Arsenic in Lake Washington, Seattle," Presented at the 37th Annual Meeting of the American Society of Limnology and Oceanography, June 23-28, Seattle, WA, 1974.

Crecelius, E. A., "Chemical Changes in Arsenic Following Ingestion by Man," Presented at the 15th Hanford Life Sci. Symp., September 29-October 1, Sponsored by ERDA (To be published in ERDA Symposium Series), 1975.

Crecelius, E. A. and R. Carpenter, "Arsenic Distribution in Waters and Sediment of the Puget Sound Region," In First Annual National Science Fond., Trace Contaminants Conf., Oak Ridge Nat'l. Lab., pp. 615-625, August 1973, 1974.

Crecelius, E. A., C. J. Johnson and G. C. Hofer, "Contamination of Soils Near a Copper Smelter by Arsenic, Antimony and Lead," Water, Air and Soil Pollution, vol. 3, pp. 337-342, 1975.

Crecelius, E. A., D. E. Robertson and L. A. Rancitelli, "The Development and Characterization of Marine Standard Reference Materials (SRM's) for Chemical Speciation Determinations," Presented at the Workshop on SRM's for Marine Science, Catalina Island, Oct. 8-9, Sponsored by National Bureau of Standards, 1975.

Hill, M. W., D. J. Eatough, L. D. Hansen, K. K. Nielson and N. F. Mangelson, "Aerosol Within a Copper Smelter Analyzed by Proton Induced X-Ray Fluorescence and Calorimetry," Presented at International Nuclear and Atomic Activation Analysis Conference and 19th Annual Meeting on Analytical Chemistry in Nuclear Technology, Gatlinburg, TN, Oct. 14-16, 1975.

Kosorok, J. R., "A Handler for the Versatec Serial Printer Interface," BNWL-B-449, September 1975.

Kosorok, J. R. (speaker) and C. A. Ratcliffe, "Memory Date Storage Via DECTape," DECUS Fall Symposium, San Diego, CA, November 5-8, 1974.

Rancitelli, L. A., J. S. Fruchter, W. D. Felix, R. W. Perkins, and N. A. Wogman, "Cosmogenic Isotope Production in Apollo Deep Core Samples," to be published in Proc. of the Sixth Lunar Conference, Geochim, Cosmochim, Acta., 1975.

Rieck, H. G., Jr., J. R. Kosorok, R. W. Perkins, and N. A. Wogman, "In-Situ X-Ray Fluorescence System Design," BNWL-B-394, Battelle-Northwest, Richland, WA, April 6, 1975.

Wogman, N. A., "Current Methods for Laboratory Analysis of Environmental Levels of Radioactivity," Presented to IEEE-Radionuclide in the Environment Measurement and Monitoring, Accepted for Publication in IEEE Proceedings, to be published June 1976.

Wogman, N. A., H. G. Rieck, Jr., and J. R. Kosorok, "In Situ Analysis of Sedimentary Pollutants by X-Ray Fluorescence," Presented to A.C.S. Mini Symposium, Walla Walla, WA, November 1974, Nucl. Inst. & Meth., vol. 128, pp. 561-568, BNWL-SA-5204, 1975.

RADIOLOGICAL AND CHEMICAL PHYSICS

- **RADIATION PHYSICS**
- **RADIATION DOSIMETRY AND RADIATION BIOPHYSICS**
- **DOSIMETRY OF PARTICULATE SOURCES IN THE LUNG**

• RADIATION PHYSICS

The radiation physics program is directed toward understanding the basic mechanism by which charged particles lose energy in traversing matter, and presenting this information in a way meaningful to the study of radiation dosimetry and biological damage. Our measurements of the absolute cross sections for the ejection of electrons from ionization by fast charged particles provide detailed information needed to test theoretical descriptions of the ionization process. Also, such measurements provide basic information required in developing particle track models for the study of energy transfer and radiation damage. The measurements of optical fluorescence from liquid systems, particularly the temporal distributions at very short times following proton and uv excitation, allow testing of theoretical models of energy transfer. We are developing such models to link our knowledge of the initial energy deposition mechanisms with the formation of early chemical species.

Our measurements indicate that electron emission cross sections from a wide range of molecules can be scaled in terms of the number of weakly bound electrons in each molecule. This information allows for ease and accuracy in utilization of analytic fits to the data for microdosimetric calculations. In addition to the studies of gas targets, we have preliminary analyses of electron emission cross sections for proton bombardment of carbon foils. These results are useful in assessment of the importance of solid state effects.

Preliminary measurements of electron ejection cross sections for energetic carbon ions indicate that simple scaling procedures are not adequate to determine ionization cross sections for heavy ions from those measured for proton impact. Compared to proton impact, ionization by energetic carbon ions results in considerably larger cross sections for production of electrons with sufficient energy to cause further ionization.

Nonexponential decay of fluorescence has been observed in both polar and nonpolar solutions. Data obtained with dilute solutions of benzene in cyclohexane have been successfully analyzed in terms of quenching by an inhomogeneous distribution of cyclohexyl radicals. A more general model is presented which accounts for production of excited states by energy transfer, LET effects and will be extended to include excitation by secondary electrons.

Electron Emission from Proton Bombardment of Molecular Targets

L. H. Toburen, W. E. Wilson,
L. E. Porter,* and D. J. Lynch**

Measurements of the energy and angular distributions of electrons ejected in ionizing collisions with fast protons have continued with emphasis on molecular targets containing low- z ($z < 10$) elements. Electron ejection cross sections for molecular targets are important to an understanding of the effects arising from the phase and molecular configuration of the media on ion track structure. In addition, these cross sections are necessary as a source term in making realistic calculations of microscopic energy deposition in biological systems since presently no experimental approach appears capable of accurately measuring the required quantities.

During the past year, we have completed our analysis of data from several hydrocarbons (methane, ethane, ethylene, acetylene, and benzene),¹ and ammonia, mono- and di-methylamine,² and acquired additional and/or new data on N_2 , N_2O , CO , CO_2 , O_2 , H_2 and H_2O . The most important observation, including the newer data, is the nearly universal applicability of scaling by the number of weakly-bound electrons in the polyatomic molecule of low- z constituents. This scalability was first observed for the group by hydrocarbons and was also found to hold for the amines and ammonia. If this scaling is indeed adequate for low- z constituent molecules, a consistent set of parameters may be attained to describe the emission cross sections and greatly simplify the use of accurate cross sections in model dosimetry calculations.

The degree of scalability found for the molecular group N_2 , N_2O , CO , CO_2 , O_2 , and H_2O is illustrated in Figure 2.1 for 1.5 MeV proton impact. With the exception of H_2O , this group of molecules is nonhydrogenous. The angular distributions for selected electron energies shown in Figure 2.1 have been scaled according to the number of weakly bound electrons in each molecule (total number of electrons

minus K-shell electrons of C, O, and N). The data for all the molecules in this group fall within the bands indicated. For comparison, the angular distributions for H_2 are indicated by the solid lines; note that the angular distributions for the nonhydrogenous group are more isotropic than are the ones for H_2 .

FIGURE 2.1. Angular Distributions for Ejection of Electrons at Selected Energies by 1.5 MeV Protons from the Group of Molecules, N_2 , N_2O , CO , CO_2 , O_2 , and H_2O . The cross sections are scaled according to the number of weakly bound electrons in each molecule.

* University of Montana, Missoula, Montana.

** Columbia Basin Community College, Pasco, Washington.

A comparison is made in Figure 2.2 of the angular distributions at 1.0 MeV for water vapor, oxygen, and hydrogen with the previously reported hydrocarbon group.¹ Water vapor and oxygen are representative of the nonhydrogenous group which was illustrated in Figure 2.1. For low-energy electron emission, the distributions for the nonhydrogenous targets are more nearly isotropic, i.e., exhibit a smaller binary-encounter peak than the distributions from hydrocarbons. At forward and back angles (and for high electron energies, at all angles), there appears to be no significant systematic differences among the distributions for hydrocarbons and nonhydrogenous targets. Note also that at forward and back angles both groups scale differently from hydrogen for all electron energies.

Distributions in energy for emission of secondary electrons, obtained by integration over angle of double-differential distributions, also exhibit some dependence on molecular structure. A comparison is made in Figure 2.3 of the energy distribution for H_2O and O_2 and the previously obtained hydrogen and hydrocarbon groups for protons of 0.3 and 1.0 MeV. The yields are expressed per electron by dividing the molecular yields by the respective number of weakly bound electrons in the target molecule. The difference between hydrogen and the hydrocarbon group is not as obvious in the energy distributions as it was in the double-differential cross sections (DDCS), because in taking the integral of DDCS to obtain the energy distribution, the yields are multiplied by the differential solid angle which, of course, includes the $\sin \theta$ term. $\sin \theta$ is small where the differences between hydrogen and hydrocarbons are greatest, hence, the differences attributed to molecular affects in the scaled DDCS do not readily show in the integrated single-differential cross sections. The situation is reversed with respect to the differences between the hydrocarbon group and nonhydrogenous group. The largest differences in the DDCS between these two groups occur where $\sin \theta$ is large; hence, in the energy distributions in Figure 2.3, the nonhydrogenous group deviates from the

FIGURE 2.2. Comparison of Angular Distributions for Ejection of Electrons of Selected Energies by 1.0 MeV Protons from H_2O and O_2 with Similar Distributions from a Group of Hydrocarbons and Hydrogen. The cross sections are scaled according to the number of weakly bound electrons in each molecule.

hydrocarbons more than does H_2 . According to Figure 2.3, there is a small but real dependence of the ejected electron spectrum below about 100 eV on molecular structure of the target, and the effect is larger for 0.3 MeV protons than for 1.0 MeV protons. We hope to investigate these molecular structure effects in more detail by careful measurements of relative cross sections for experimental conditions which enhance the target-dependent differences.

FIGURE 2.3. Energy Distributions of Secondary Electrons Ejected by 0.3 and 1.0 MeV Protons from Various Targets. The cross sections are scaled according to the number of weakly bound electrons in each molecule. The solid line represents the average scaled cross sections for the hydrocarbons investigated by Wilson and Toburen.¹

Electron Emission from Proton Bombardment of Carbon Foils

L. H. Toburen, H. Paretzke,* and W. E. Wilson

It is not generally feasible to make direct measurements of ionization cross sections in a condensed media since the product ions and electrons interact with the media before they can be collected. Still, it is the

condensed media where biological damage occurs; how then can we determine to what extent gas phase data are usable in predicting interactions which take place in solid or liquid media? We are presently investigating this question in collaboration with Dr. H. Paretzke of the Institute of Strahlenschutz, Neuherberg (Munich), Germany. Dr. Paretzke is using our gas phase data as input to a Monte Carlo code which will be used to calculate the spectra of electrons ejected from a thin foil by fast proton impact. These calculated multiple collision spectra will then be compared to measured cross sections for ejection of electrons from thin carbon foils. We hope in this way to determine whether gas phase collision data are adequate to predict the results obtained from the condensed system or whether solid state effects, such as plasmon excitation, must be included.

Calculations by Dr. Paretzke have not been completed, however, preliminary measurements of electrons ejected from a carbon foil have been made. A comparison of the cross sections, per weakly bound electron (principle quantum number 2), for gas and foil targets is shown in Figure 2.4. The foil spectra are, of course, quite different from those for gas targets due to the high probability of multiple scattering of high energy electrons within the foil. This scattering results in relatively larger cross sections for observing high energy electrons at the larger angles (angles smaller than 50° have not yet been measured for foil targets). Note particularly the large cross section for ejection of electrons with energies as large as 1300 eV at 125° for the foil target compared to a negligible cross section for ejection of electrons from the gas target for electrons with energies greater than 500 eV. Those electrons observed at large angles for the foil are presumed to arise from the scattering of electrons initially ejected in the forward direction; note the large cross section for ejection of fast electrons in the 15° gas target data. We expect that similarly large cross sections will be obtained from the results of a Monte Carlo code which takes into consideration large angle and multiple scattering of electrons within the foil. At the low energy end of the ejected electron energy spectrum, the foil target cross

* Institut für Strahlenschutz, Neuherberg, Germany.

FIGURE 2.4. Cross Section Per Target Electron for Electron Ejection from Methane Gas and $3.0 \mu\text{g}/\text{cm}^2$ Carbon Foil by 1.0 MeV Protons

sections are somewhat smaller than the gas phase results. This may be expected since the low energy electrons generated within the foil have ranges shorter than the foil thickness ($3 \mu\text{g}/\text{cm}^2$ in the case illustrated in Figure 2.4) and cannot reach the detector. Only the electrons ejected at or near the surface with these low energies are detected.

From the qualitative comparison of gas and foil data, there appears to be no obvious structures observed in the foil data that cannot be predicted by multiple scattering of gas phase data. We await the results of the Monte Carlo code to determine whether

new features are evident when a quantitative comparison is made.

Consistency Tests of the Measured Cross Sections

L. H. Toburen and S. T. Manson*

Recently, Y.-K. Kim, Argonne National Laboratory, has presented a theoretically based consistency test to aid in determining the reliability of measured differential ionization cross sections.³ This procedure involves the use of the Rutherford

* Georgia State University, Atlanta Georgia.

theory (Mott theory for electron impact) to obtain cross sections for ejection of relatively high energy electrons and the use of measured oscillator strength distributions to estimate the shape of the low energy portion of the ejected electron spectra. If these two ends of the spectrum can be estimated in this manner, it is then a relatively simple matter to obtain the whole energy spectrum by making a smooth transition at intermediate energies. The success of this method was illustrated by Kim in an application to cross sections for incident electrons with energies of 500 eV.³ The obvious extension of this work to proton impact is to be considered here. The primary questions to be answered regarding the application of this technique are first, how high in energy must the particles be for these approximations to be useful, and second, how important are inner shell effects where the target atom has a relatively complex electronic structure.

First we explore the consistency test for high energy ejected electrons. For proton impact, the Rutherford cross section can be written as

$$\frac{d\sigma}{dE} = \frac{4\pi A_0^2 z^2 R^2}{T} \sum_j \frac{N_j}{E_j^2} \quad (1)$$

where z is the charge of the incident particle, R is the Rydberg unit of energy, A_0 is the Bohr radius of the hydrogen atom, T is the kinetic energy of the proton in units of equivalent velocity electron energy ($1/2 m_e v_p^2$), E_j is energy lost in ionizing an electron which is initially in the j th electronic level ($E_j = I_j + \epsilon$, where ϵ is the ejected electron energy and I_j is the binding energy of the initial state), and N_j is the number of electrons in the j th electronic state. If we plot the quantity $Y(E)$, defined as

$$Y(E) = \frac{TE^2}{4\pi A_0^2 z^2 R^2} \sigma(E) \quad (2)$$

where $\sigma(E)$ is the measured single differential cross section and $E = I + \epsilon$ with I now defined simply as the first ionization potential, as a function of E , then the high energy portion of the ejected electron spectrum should ap-

proach a constant value equal to the number of electrons in the atom. This is illustrated in Figure 2.5 for proton ionization of helium where indeed, for the higher energy protons, the ejected electron spectra approaches a constant value which is approximately two (the number of electrons in the helium atom). The measured spectrum actually approaches a value 10% larger than the Rutherford value; this may be evidence of a systematic 10% error in our measurements, however, further analysis makes this less clear. For example, the open square at zero ejected electron energy is considered a reasonably accurate calculation of the zero energy cross section. This value was obtained by Kim et al.,⁴ based on extrapolation of the discrete excitation cross sections into the continuum. Since our results appear to agree well with this point for 1 MeV, where time-of-flight measurements are incorporated to improve our low energy accuracy, it is hard to understand a systematic 10% error. The possibility of an ejected electron energy dependent error does exist, but it is not yet clear whether this evidence is sufficient to be convincing. The 1.5 and 0.3 MeV results have not been updated at low energies with TOF data, therefore, cross sections below 30 eV have larger uncertainties. (The arrows in Figure 2.5 are positioned at 30 eV ejected electron energy.) In any case, the Rutherford cross section appears a good guide for emission cross sections for ejected electron energies greater than about 30 Rydbergs and proton energies of 1.0 MeV and above for a simple target such as helium.

If we consider more complex targets, the conclusions are even less obvious. Differential ionization cross sections for proton ionization of argon are shown in Figure 2.6. For this atom with a filled $n = 3$ shell, it is not clear just what N should be used in the Rutherford formula. It appears that the cross sections for 1.0 and 1.5 MeV protons level off to a value of about 13, indicating according to the Rutherford estimate, that 13 electrons participate in the collision process. By defining $Y(E)$ in the manner of Eq. (2) and taking the value of I to be equal to the first ionization potential, the Rutherford value is no longer a constant as inner shells

FIGURE 2.5. Ejected Electron Energy Distributions for Proton Ionization of Helium. The scaling parameter $Y(E)$ is defined in Eq. (2). The arrows indicate an electron energy of 30 eV and the open square at $E/R = 1.81$ are calculated values for ejection of zero energy electrons obtained from Y.-K. Kim, et al.⁴

FIGURE 2.6. Ejected Electron Energy Distributions for Proton Ionization of Argon. The scaling parameter $Y(E)$ is defined in Eq. (2). The arrows represent an electron energy of 30 eV and the open square at $E/R = 1.15$ are calculated values from Y.-K Kim, et al.⁴

become ionized owing to the inner shell contribution to the N_j/E_j term. The fact that inner shells are ionized is apparent in that the L-Auger peak at about 15 Rydbergs is relatively intense. This analysis technique must, therefore, await more sophisticated theoretical treatment to account more accurately for the inner shell contributions.

A second type of plot which Kim emphasizes for use in arriving at accurate low energy cross sections is illustrated in Figure 2.7 for proton ionization of argon. In this case, $Y(E)$ is plotted versus R/E which emphasizes the low energy portion of the spectrum and presents the differential cross section in such a way that the area under the curve is proportional to the kinetic energy of the proton times the total ionization cross section. The square symbol plotted at $R/E = 0.86$ in Figure 2.7 is the cross section for ejection of zero energy electrons calculated using the parameters of Kim, et al.⁴ The dashed curves are derived from measured oscillator strength distributions presented by Kim and normalized at $\epsilon = 0$ to the calculated zero energy cross section value. Based on Born theory, the oscillator strength distribution should characterize the shape of the differential ionization cross section curve for low energy ejected electrons if the incident charged particle is of sufficiently high energy. The question remains as to what is sufficiently high energy when the incident charged particle is a proton. The results shown in Figure 2.7 for 1 MeV protons include time-of-flight measurements and should be accurate to 20% to electron energies as low as 1 eV, $R/E = 0.82$. The results shown for 0.3 and 1.5 MeV do not yet include time-of-flight results. These results are less reliable for electron energies below 20 eV, $R/E = 0.4$. Even though the measurements at 0.3 and 1.5 MeV haven't been updated at low electron energies using TOF techniques, there is qualitative evidence that agreement between the oscillator strength distributions and differential ionization cross sections is better as proton energies increase and that 1.5 MeV is still somewhat inadequate as a representation of the high energy approximation.

FIGURE 2.7. Ejected Electron Energy Distributions for Proton Ionization of Argon. The dashed lines represent measured oscillator strength distributions ($E df/dE$) normalized to the calculated zero energy cross section value. The oscillator strength distributions and zero energy cross sections were obtained from Y.-K. Kim, et al.⁴

In the region of the Cooper minimum ($R/E \approx 0.3$), the numerous additional ionization channels excited by protons, compared to photon or electron ionization lead to a much smaller minimum in the value of $Y(E)$. As was observed for the comparison of cross section for energetic ejected electrons to Rutherford cross sections, this analysis lends itself much better to simple target systems. Differential cross sections of proton ionization of neon are shown in Figure 2.8. These results show excellent agreement between oscillator strengths and ionization cross sections for electron energies from 10 to 50 eV and protons of 1.5 MeV. The addition of TOF measurements to the 1.5 MeV cross

FIGURE 2.8. Ejected Electron Energy Distributions for Proton Ionization of Neon. The triangular and square points are zero energy ejected electron cross sections for 1.5 and 1.0 MeV, respectively. The dashed line represents measured oscillator strength distributions ($E \frac{df}{dE}$) normalized to the zero energy point for 1.5 MeV proton impact. The zero energy points and oscillator strength distributions were obtained from Y.-K. Kim, et al.⁴

sections should improve agreement for electron energies below 10 eV. It should be noted that the 1 MeV results, which do include TOF measurements, do not have the proper shape at low energies. We believe this to be evidence that 1 MeV is not sufficiently high energy for the dipole approximation to be useful. For electron energies above 50 eV, the dipole oscillator strength is inadequate because of numerous continuum channels contributing to the ionization process for incident protons. These channels are not all available in the photo-ionization process.

We are presently extending this work to include time-of-flight measurements to complement the present results of electrostatic analysis and hope that more definitive statements regarding the Kim analysis technique can then be made.

Ionization in Heavy Ion Collisions L. H. Toburen and W. E. Wilson

In the investigation of energy deposition by fast charged particles

and/or neutrons, one must consider the contributions of fast heavy recoil ions. Although these ions have extremely short ranges and normally are assumed to deposit their energy locally, the electrons generated in ionizing collisions may transfer some portion of the collision energy from the local site. In addition to questions regarding the dimensions over which the energy is deposited, it is also necessary to know the number and energy of electrons generated through ionization to determine whether further ionization will result from these electrons. We have, therefore, made a preliminary measurement of the energy and angular distributions of electrons ejected in carbon ion collisions with argon, methane, and molecular hydrogen to gain some insight into the heavy ion collisions. Such data are required to provide reliable model calculations of track structure and, thereby, accurate assessments of the effects of heavy ion collisions in dosimetric determinations.

Measurements of electron emission from an argon target by a singly

ionized carbon ion beam at 1.3 MeV are shown in Figure 2.9. These results are compared to similar measurements for incident protons of approximately the same energy (1 MeV) and of nearly the same velocity (0.1 MeV).⁵ Recall that in a plot of this form, $Y(E)$ versus R/E , the area under the curve is proportional to the kinetic energy of the incident ion times the total ionization cross section. In the case illustrated in Figure 2.9, we have taken the incident ion charge of plus one to be the z value of the incident particle in evaluating $Y(E)$ (note that, as defined in Eq. (2) of the last section, $Y(E)$ should include the projectile charge in the denominator). By integration, one finds the total cross section for ionization by C^+ is more than an order of magnitude greater than for ionization by protons of similar energy, $1.95 \times 10^{-15} \text{ cm}^2$ for C^+ compared to $1.25 \times 10^{-16} \text{ cm}^2$ for H^+ , and approximately a factor of 4 greater than ionization by protons of similar velocity, $5.17 \times 10^{-16} \text{ cm}^2$. We do not place any particular significance to this type of comparison except to note the large magnitude of the ionization cross section for carbon ions and to indicate that simple scaling of the cross sections based on the charge of the incident particle, such as setting z equal to 1 to imply

a totally screened nucleus or z equal to 6 to imply no screening, is completely inadequate.

A somewhat more startling result appears to be the energy of the electrons which contribute the major portion of the ionization. Whereas the major contribution to the area of the curve in Figure 2.9 for incident protons is for electrons with energies less than about 50 eV, a large contribution for carbon ion impact occurs for electron energies above 50 eV and the peak in the $Y(E)$ distribution occurs at about 200 eV. The implications of this result are that a large fraction of the electrons ejected by carbon ion impact are sufficiently energetic to produce further ionization and that these electrons may transfer energy out of small sensitive sites. It should also be pointed out that Figure 2.9 provides a clear illustration of the inadequacy of using simple scaling techniques to obtain cross sections for heavy ions from our knowledge of similar cross sections for protons.

Some of the structural features of the ionization process of heavy ion impact are shown in the ejected electron energy spectra shown in Figure 2.10. These spectra for ionization of

FIGURE 2.9. Ejected Electron Distributions for Ionization of Argon by 1.3 MeV C^+ Ions and by Protons of Nearly the Same Energy (1.0 MeV) and Velocity (0.1 MeV). The 0.1 MeV results are from Crooks and Rudd.⁵

FIGURE 2.10. Double-Differential Cross Sections for Electron Ejection from CH_4 and H_2 by 1.3 MeV C^+ Ion Impact

hydrogen and methane by 1.3 MeV C^+ show Auger groups associated with inner shell ionization of carbon. The methane spectrum shows Auger lines originating from K-shell ionization of carbon in the methane target as well as K-shell ionization of the incident carbon ion. Since the Auger electrons ejected from the projectile come from a moving reference frame, their energy is Doppler shifted and observed at about 320 eV in the 60° spectrum shown in Figure 2.10, in contrast to the peaks near 240 eV which are Auger transitions in the stationary target. By determining the total area under the Auger peaks (one must include corrections to account for changes in the solid angle for Auger emission from a solid angle of the projectile as viewed in the laboratory rest frame), the total cross section for K-shell ionization of carbon in C^+ -methane collisions can be obtained. The value obtained in these preliminary measurements ($\sigma_K = (6 \pm 2) \times 10^{-18} \text{ cm}^2$) is in excellent agreement with the x-ray measurements of Der, et al.⁶ for C^+ on solid carbon if a fluorescence yield of 2.2×10^{-3} is used to arrive at the ionization cross section from the measured x-ray yield. This fluorescence yield is the most recent published value for 0.1 to 1.0 MeV proton ionization of both solid and gaseous (CH_4) targets.⁷

If we go one step further and determine the K-shell ionization cross

section for the projectile in C^+ -hydrogen collisions, one might be tempted to compare this value with the collision in reverse order, i.e., protons on carbon. To make such a comparison, consider the molecular hydrogen target to be equivalent to two protons. We then find that one-half the measured cross section for ionization in C^+ collisions with molecular hydrogen ($3.2 \times 10^{-19} \text{ cm}^2$) compares amazingly well with the value of 1.5×10^{-19} obtained for proton impact ionization of the K-shell of carbon in a carbon target,⁸ where again fluorescence yield of 2.2×10^{-3} was used for the fluorescence yield of carbon. It should be noted that these comparisons are made for scaled particle energies, i.e., units of keV/amu. It is a bit dangerous to base conclusions on these preliminary data, however, it appears from this excellent agreement with previous x-ray measurements, that the fluorescence yield for carbon is nearly independent of whether the determination is for ionized carbon (C^+), gaseous (CH_4), or solid carbon targets.

Pulsed Radioluminescence Studies J. H. Miller and M. L. West

Pulsed radioluminescence studies continue to emphasize the importance of fluorescence response as a useful technique for probing early physical-chemical effects of ionizing radiation. Excited molecules serve as sensitive

probes through their interaction with the surrounding environment via energy transfer and quenching mechanisms. The use of pulsed excitation techniques and single photon counting permits reliable measurements of small populations of excited states on the nanosecond time scale. These data provide sensitive tests for theoretical models being developed to explain the evolution of chemically active species originating from primary energy deposition events in the physical stage.

The interpretation of fluorescence data from dilute solutions of benzene in cyclohexane is simplified by the predominance of one radical, C_6H_{11} , and the absence of a long-lived fluorescing component.⁹ We have developed a model to explain the nonexponential decay of fluorescence following proton irradiation which is based on solution of the coupled diffusion equations:

$$\frac{\partial S^*(\vec{r}, t)}{\partial t} = D\nabla^2 S^*(\vec{r}, t) - k_{uv} S^*(\vec{r}, t) - k_{R*} S^*(\vec{r}, t) R(\vec{r}, t) \quad (1)$$

$$\frac{\partial R(\vec{r}, t)}{\partial t} = D\nabla^2 R(\vec{r}, t) - k_{RR} R^2(\vec{r}, t) \quad (2)$$

The first equation describes the change in concentration of excited benzene molecules due to diffusion, uv-type decay modes and radical quenching. The concentration of radicals is determined from Eq. (2), where k_{RR} is the rate constant for radical-radical recombination. The diffusion coefficient is approximately 2×10^{-5} cm²/sec. These equations do not account for the formation of radicals and excited states, which are the results of rapid relaxation phenomena occurring on a picosecond time scale after irradiation. The number and spatial distribution of chemical species resulting from these phenomena enter the model as a boundary condition on Eq. (1) and (2) at $t = 0$.

Methods for solving diffusion equations like (1) and (2) have been reviewed by Kupperman.¹⁰ For this analysis, we have adapted a method proposed by Ganguly and Magee.¹¹ The chemical species originating from an energy deposition event in the physi-

cal stage are assumed to form a cluster about a point, $\vec{r}_i$. At time t , the cluster contains $n_{Ri}(t)$ and $n_{*i}(t)$ radicals and excited states which are Gaussian distributed about $\vec{r}_i$ with a width $2D\tau_i = 2Dt + a_i^2$. We shall refer to a_i as the cluster radius at $t = 0$. The concentration of excited states and radicals are a superposition of these clusters,

$$S^*(\vec{r}, t) = \frac{\sum_{i=1}^N n_{*i} \exp \left[- \left(\vec{r} - \vec{r}_i \right)^2 / 4D\tau_i \right]}{(4\pi D\tau_i)^{3/2}} \quad (3)$$

$$R(\vec{r}, t) = \frac{\sum_{i=1}^N n_{Ri} \exp \left[- \left(\vec{r} - \vec{r}_i \right)^2 / 4D\tau_i \right]}{(4\pi D\tau_i)^{3/2}} \quad (4)$$

When Eq. (3) and (4) are substituted into Eq. (1) and (2) and the integral over $\vec{r}$ is performed, we obtain differential equations for the stochastic processes $n_*(t)$ and $n_R(t)$. From the solution of these equations, we obtain information about the probability distribution of n_{*i} and n_{Ri} at time t given information about the distribution of the parameters $\vec{r}_i$, a_i , and $n_{Ri}(0)$. If statistical fluctuations in the number and distribution of chemical species in clusters are ignored, these equations take the form

$$\frac{d}{dt} \ln \bar{n}_* + k_{uv} + k_{R*} \bar{n}_R Q / (8\pi D\tau)^{3/2} = 0 \quad (5)$$

$$\frac{d}{dt} \bar{n}_R + k_{RR} \bar{n}_R^2 Q / (8\pi D\tau)^{3/2} = 0 \quad (6)$$

where $\tau = t + r_o^2/2D$, and

$$Q = \left\langle \frac{1}{N} \sum_{i=1}^N \sum_{j=1}^N \exp \left[- \left(\vec{r}_i - \vec{r}_j \right)^2 / 8D\tau \right] \right\rangle \quad (7)$$

The functions $\bar{n}_*(t)$ and $\bar{n}_R(t)$ denote the mean number of excited states and radicals per cluster, and r_o is the RMS initial cluster radius.^o

The evaluation of Q requires a track model. If we consider only excitation in the track core, and assume the clusters are centered on the track axis, then Q is approximated by

$$Q = 1 + (8\pi D\tau)^{1/2}/d \quad (8)$$

where d is the mean separation between cluster centers.¹¹ In this approximation, Eq. (5) and (6) have the solution

$$\bar{n}_*(t) = \bar{n}_*(0) \exp(-k_{uv}t)$$

$$\left[1 + U_\alpha(t) + U_\beta(t) \right]^{-k_{R^*}/k_{RR}} \quad (9)$$

$$\bar{n}_R(t) = \bar{n}_R(0) \left[1 + U_\alpha(t) + U_\beta(t) \right]^{-1} \quad (10)$$

$$\text{with } U_\alpha = \frac{\bar{n}_R(0) k_{RR}}{8\pi Dd} \ln \left(1 + \frac{2Dt}{r_o^2} \right) \quad (11)$$

$$U_\beta = \frac{\bar{n}_R(0) k_{RR}}{8\pi D\pi^{1/2} r_o} \left[1 - \left(1 + \frac{2Dt}{r_o^2} \right)^{-1/2} \right] \quad (12)$$

U_β is the contribution from radical recombination within a cluster, and U_α is the contribution from intercluster recombination. The radical recombination rate k_{RR} is estimated to be $2.5 \times 10^{-9} \text{ M}^{-1}\text{sec}^{-1}$ on the basis of a measured yield of 5 radicals per 100 eV.¹² The uv decay rate, k_{uv} , is measured for each sample in a separate experiment.¹³ By fitting the fluorescence profile observed under proton irradiation, we obtain estimates of the dimensionless ratios

$$q = k_{R^*}/k_{RR},$$

$$\alpha = \bar{n}_R(0) k_{RR}/8\pi Dd,$$

$$\beta = \bar{n}_R(0) k_{RR}/8\pi D\pi^{1/2} r_o, \text{ and}$$

$$\gamma = 2D/r_o^2 k_{uv} \quad (13)$$

From these ratios, we can determine k_{R^*} , r_o , d , and $\bar{n}_R(0)$ in terms of k_{uv} , k_{RR} , and D . This procedure requires an accurate determination of β which is possible only when recombination within clusters competes with recombination from overlapping clusters. When this is not the case, only the ratio $\bar{n}_R(0)/d$ can be determined from the data. This ratio may also be written as $\bar{S}_C/\epsilon_R$, where $\bar{S}_C$ is the energy loss per unit length in the core averaged over proton energies less than the initial energy, E_p , and ϵ_R is the mean energy loss per radical formed (i.e., $\epsilon_R = 100 \text{ eV/g}_R$). Figure 2.11 shows the variation of $\bar{S}_C$ with initial proton energy.¹⁴ Preliminary results based on data taken at 1.7 MeV initial proton energy are shown in Figure 2.12. Values obtained for the parameters are listed in Table 2.1. Note that due to the temperature change, the uv lifetimes differ by a factor of three. Differences in the calculated values of k_{R^*} , r_o , and g_R probably reflect the uncertainty in the parameters q , γ , and α rather than temperature variation.

The parameter β cannot be determined from this data. If we assumed that 100 eV is the mean energy absorbed per cluster, then the data predicts a value of 40 Å for the mean distance between cluster centers. If this is the case, we have strongly overlapping clusters at all times (i.e., cylindrical geometry is a good approximation). Equation (8) can be used to establish an approximate criterion for observation of a spherical component of radical recombination. If $d = (8\pi D\tau)^{1/2}$, then intra- and intercluster interaction have equal importance. If d is inversely proportional to the averaged stopping power, $\bar{S}_C$, then intracluster recombination should contribute about one part in three at 4 MeV and 1 nsec. With improved time resolution and

good counting statistics, this should permit an estimate of β . We emphasize again that determination of β is important because it allows us to calculate both the track parameters $n_R(0)$ and d without making the artificial assumption of 100 eV mean energy absorbed per cluster.

FIGURE 2.11. Variation of the Averaged Stopping Power in the Track Core with Initial Proton Energy, E_p

FIGURE 2.12. Least Squares Fit to the Fluorescence Decay of Benzene in Cyclohexane Following Irradiation by 1.7 MeV Protons

TABLE 2.1. Results of Least Squares Analysis of Non-Exponential Fluorescence Decay(a)

$T(^{\circ}\text{C})$	$k_{uv}^{-1}(\text{nsec})$	$k_{R*}(10^{10}\text{M}^{-1}\text{sec}^{-1})$	$r_o(\text{\AA})$	g_R	$\chi_v^2(b)$
14	36	1.5	34	2.8	1.02
49	13	1.2	29	2.4	1.22

a. Calculations based on $k_{RR} = 2.5 \times 10^9 \text{M}^{-1}\text{sec}^{-1}$,
 $D = 2 \times 10^{-5} \text{cm}^2/\text{sec}$, and $\bar{S}_C(1.7 \text{ MeV}) = 2.5 \text{ eV/\AA}$.
 b. Chi square per degree of freedom.

The results presented in Table 2.1 illustrate the type of information which can be obtained from this analysis of the time resolved fluorescence data. The uncertainty in the parameters can be substantially reduced by improvements in time resolution and counting statistics. Extensions of the model to account for statistical fluctuations in the number and distribution of chemical species in a cluster, displacement of the cluster centers perpendicular to the track axis, and δ -ray effects are also being investigated.

Quenching of excited states by radicals reduces the energy output from the liquid in the form of uv fluorescence. Consequently, the model which we have developed to analyze the fluorescence decay rate can also be used to predict the α/β ratio, if we assert that the reduction in light output with high LET radiation is due primarily to the more efficient quenching of excited states when clusters are strongly overlapping. We define the α/β ratio as the ratio of the time integrated fluorescence intensity after absorption of the same amount of energy in the form of high and low LET radiation. If we assume that the number and distribution of radicals and excited states within a cluster are the same for high and low LET radiation, then our model predicts

$$\frac{E_f^\alpha}{E_f^\beta} = \frac{\int_0^\infty e^{-k_{uv}t} \left[1 + U_\alpha(t) \right]^{-k_{R^*}/k_{RR}} dt}{\int_0^\infty e^{-k_{uv}t} \left[1 + U_\beta(t) \right]^{-k_{R^*}/k_{RR}} dt} \quad (14)$$

where U_α and U_β are defined in Eq. (11) and (12). Figure 2.13 shows the change E_f^α/E_f^β with $\bar{S}_C$ for fixed values of r_0 , $n_R(0)$, and k_{uv} . This suggests that measurements of the α/β ratio for dilute solutions would provide additional tests of our model.

Experimental measurements of fluorescence response curves for protons and uv irradiation of benzene in polar and nonpolar solvents are in progress. Typical decay curves for proton irradiation are shown in Figure 2.14. In both polar and nonpolar solvents, the

FIGURE 2.13. Predicted Change in α/β Ratio with Averaged Stopping Power Based on Radical Quenching Model

FIGURE 2.14. Fluorescence Decay for Excitation by 1.7 MeV Protons

fluorescence decay is nonexponential for proton irradiation and the rate of decay [defined as $d(\ln I/I_0)/dt$] approaches that observed for uv excitation. The dashed lines in Figure 2.14 indicate the measured rate of decay for uv excitation (k_{uv}). These response curves imply an initial rapid quenching that disappears in time to yield the exponential decay observed for uv excitation. These data will provide additional tests of the recently developed diffusional quenching model. Measurements in methanol, ethanol, and water will be used to investigate the effects of solvated electrons on fluorescence quenching. For these systems, it is anticipated that a more general picture of multiradical quenching will be needed.

Fluorescent lifetime measurements as a function of solvent temperature are in progress. Our previous studies¹³ of the temperature dependence of benzene fluorescence suggests that the solvent merely acts as an inert media to furnish a rapid thermal relaxation of the initially populated levels. Decay rates in solution are thus described by

$$k = k_0 + k_1 e^{-W/KT}$$

where k is related to intramolecular process and $k_1 e^{-W/KT}$ describes the thermally activated quenching process. In order to reliably determine W , fluorescence lifetimes must be measured over a wide range of solvent temperatures. Our present pulsed uv system does not accurately measure short lifetimes ($\tau < 10$ nsec). A new pulsed uv system with a 1 nsec response is being assembled and will allow us to make measurements at the higher temperatures where fluorescence lifetimes are typically a few nanoseconds.

The previous analysis of a nonexponential decay in solutions of benzene in cyclohexane under proton irradiation neglected the role of energy transfer from excited cyclohexane molecules. However, there is experimental evidence¹⁵ that such energy transfer does occur from a short-lived excited state. If we assume an initial excited state population of benzene, S_0^* , and cyclohexane, C_0^* , and if

cyclohexane transfers energy to benzene at the rate k_{CS} to create additional excited molecules, then

$$S^*(t) = S_0 e^{-G(t)} \left\{ 1 + \frac{k_{CS} C_0^*}{k_C + k_{CS} S_0 - k_S} \cdot \left[1 - e^{-(k_C + k_{CS} - k_S)t} \right] \right\} \quad (15)$$

where k_S and k_C are the uv decay rates of benzene and cyclohexane, respectively. There is an initial rapid rise determined by the lifetime of excited cyclohexane and for times greater than 1 nsec, the term $e^{-(k_C + k_{CS} - k_S)t}$ is negligible and the decay reduces to $Ae^{-G(t)}$, where $G(t)$ is the nonexponential decay of benzene fluorescence currently observed with nanosecond resolution and A is determined from Eq. (15).

The rise time due to energy transfer has a characteristic width

$$t = \frac{1}{k_C + k_{CS} S_0 - k_S} \sim 200 \text{ psec} \quad (16)$$

for $S_0 = 0.01M$ and with values of k_C and k_{CS} reported by Beck and Thomas.¹⁶ If a system response of this magnitude (~ 200 psec) is experimentally feasible and if simultaneous measurements of this response with the fluorescence response can be made, least square convolution of system response with Eq. (15) should permit us to test the validity of this model and to obtain estimates of its parameters. A program to perform these operations is being developed and will be initially tested using synthetic data generated from Eq. (15) and Gaussian distributed errors.

We have considered in detail the experimental feasibility of obtaining a system resolution of ~ 200 psec. The minimum proton pulse resolution depends on beam optics design. There is at present no convenient method of accurately measuring this subnanosecond proton pulse shape. Techniques using a sampling scope and Faraday cup are limited by the small amount

of current per proton burst. A technique using secondary electron emission for current amplification is being considered and needs to be tested in the lab. Both theoretical calculations and duty cycle measurements indicate a practical limit of ~ 200 psec FWHM for the proton beam width.

The major unresolved problem for subnanosecond measurements is the de-

termination of the detector-proton beam response function, both in shape and time relative to an observed fluorescence response curve. At present, we are investigating two experimental techniques that may solve this problem: 1) a fast decaying organic dye ($\tau \sim 100$ psec), and 2) the production of Cerenkov radiation via an intermediate (p, γ) reaction.

• RADIATION DOSIMETRY AND RADIATION BIOPHYSICS

The joint purpose of these two closely integrated programs is to explore the connections between the primary physical events produced by radiation and their biological consequences in cellular systems. The radiation dosimetry program includes theoretical description of the primary events and their connection with the observable effects and experimental realization and measurement of the physical parameters needed by the theory. The radiation biophysics program tests or makes use of the theoretical developments and gathers information needed for the theories by experiments on cellular systems. This year the study of division delay in Chinese hamster ovary (CHO) cells led to identification of three groups of cells near the end of the cell cycle and the measurement of the probability of delay as a function of dose in one of the groups. Our past theoretical work provided a basis for interpreting the probability of delay. Hydroxyurea was found to slow the progress of G₂ CHO cells, a fact important in explaining the observed action of the drug. Theoretical and experimental work on Chlamydomonas reinhardtii combined to explain its response to changes in dose rate. Cross section data obtained under the Radiation Physics program were approximated by relatively simple formulae so the data could be used in Monte Carlo simulations. A theory of the microdosimetry of internal sources was developed to form the basis of a new program, Dosimetry of Particulate Source in the Lung, and some initial experimental work was done to support the theory.

Division Delay After Low X-Ray Doses
and the Effect of Cycloheximide
M. H. Schneiderman, L. A. Braby,
and W. C. Roesch

In search of a simple system in which to study the connection between primary radiation events and observable biological phenomena, radiation-induced delays were investigated. Because delays occur immediately after irradiation and because the delay in

each part of the mitotic cycle can be identified, we believe division delay may be a useful biological end point. The high sensitivity of the mitotic-selection method of measuring delays was used to study very low x-ray doses, with and without cycloheximide present. A group of cells late in the mitotic cycle played an important role in the results; therefore, their properties were also studied.

The study consisted of a number of experiments that differed only in the x-ray and in the presence or absence of cycloheximide. Log-phase monolayers of Chinese hamster ovary cells were shaken for 10 sec every 10 min and each time the mitotic cells shaken loose were collected and counted. Immediately after the tenth shake, the cultures were treated with 7-rad/min, 1.5 MV x rays or 10- μ g/ml cycloheximide. The number of cells collected on subsequent shakes was normalized to the average of the ten pretreatment shakes.

Figure 2.15 shows the normalized yields of mitotic cells for cycloheximide alone and for cycloheximide and a large x-ray dose, 150 rad. All large doses, those above about 100 rad, give curves indistinguishable from the one for 150 rad.

The difference between the two curves in Figure 2.15 is the mitotic yield of what we call the "CX cells." This group of cells is defined by the effect of cycloheximide and x rays on the progress of its members toward division. After a point late in G₂, a cell's progress is no longer affected by cycloheximide; after a later point, early in M, it is no longer af-

ected by radiation.¹⁷ The CX group comprises those cells that have passed the first of these points but not the second. Figure 2.16 shows the CX cell yield. It also shows the yield of two complementary groups: 1) the "refractory cells," those not affected by either cycloheximide or x rays, and 2) the "sensitive cells," those affected by both cycloheximide and x rays.

Figure 2.17 shows the measured mitotic yields for low x-ray doses, 4.5 to 34 rad. The clearly visible effect at 4.5 rad shows the high sensitivity of the cells to radiation. The yields at high doses are similar to these but actually reach zero for periods depending on the dose.

Figure 2.18 shows the contributions of the refractory, CX, and the sensitive cells to the yields in Figure 2.17. They show that the refractory cells were not affected by the x rays, that a dose-dependent fraction of the CX cells were not affected, and that all the sensitive cells were delayed about 10 min and then began to arrive at reduced, dose-dependent rates. We do not presently know the eventual fate of the CX cells that were delayed; we currently

FIGURE 2.15. The Mitotic Yield of Chinese Hamster Ovary Cells Treated with Cycloheximide (Δ) or with 150 Rad of X Rays in the Presence of Cycloheximide ($\circ$)

FIGURE 2.16. The Mitotic Yield of Chinese Hamster Ovary Cells Belonging to One of Three Groups: 1) "Refractory" Cells, 2) "CX" Cells and 3) "Sensitive" Cells

FIGURE 2.18. The Effect of Low Doses of X Rays on the Mitotic Yields of the Three Groups of Chinese Hamster Ovary Cells Described Under Figure 2.16

FIGURE 2.17. The Effect of Low Doses of X Rays on the Mitotic Yield of Chinese Hamster Ovary Cells

think they were delayed for longer than the period of the experiment, but they may be included with the latest sensitive cells we measured.

Figure 2.19 shows the low-dose mitotic yields of cells containing cycloheximide, and Figure 2.20 shows

the contributions to them of the refractory and CX cells--the sensitive cells do not contribute because they are stopped by the cycloheximide. Comparison of the CX contributions, Figures 2.18 and 2.20, indicates that a larger fraction of CX cells is delayed when cycloheximide is present.

FIGURE 2.19. The Effect of Low Doses of X Rays in the Presence of Cycloheximide on the Mitotic Yield of Chinese Hamster Ovary Cells

FIGURE 2.20. The Effect of Low Doses of X Rays in the Presence of Cycloheximide on the Mitotic Yields of the Groups of Chinese Hamster Ovary Cells Listed Under Figure 2.16

Figure 2.21a shows the fractions of the CX cells collected, with and without cycloheximide, as functions of the dose. The cycloheximide makes a big difference: without it the fractions form a straight line; with it, a curve. Some understanding of the reasons for these shapes can be found by comparing them with radia-

tion survival curves. The fractions can be analyzed with the same concepts and methods as survival curves because both collection and survival depend on the probabilities of single charged particles or pairs of charged particles producing events in small biochemical entities.

In earlier work,¹⁸ we suggested replotting survival data in the form $-\ln(S)/D$ versus D . Figure 2.21b shows the fractions, now denoted by S , plotted this way. The theory indicates what kinds of curves should fit the data, but here the experimental uncertainties prevent a unique fitting. We present two possibilities in Figure 2.21 that we think bracket the correct interpretation. The dotted lines represent a model in which no delays are initiated by single charged particles; only by pairs of charged particles; in this model, the mean recovery time has to be about 1 min to fit the data. The solid lines represent a model in which both single charged particles and pairs of them initiate delays; the mean recovery time has to be short--a 1/10 min or less, possibly much less. Other possible models incorporate intermediate degrees of single-particle initiated delays and of mean recovery time.

Within this range of possible interpretations, the role of cycloheximide is the same: It mimics a high dose rate. High dose rates minimize recovery by reducing the time between pairs of events. Cycloheximide, on the other hand, must minimize recovery chemically--possibly by interfering with protein synthesis.

The CX cells are much more apt to be delayed than to suffer reproductive death; the parameters for the curves in Figure 2.21 are some hundreds of times larger than the corresponding ones for reproductive survival.

Application of Kellerer's¹⁹ relation between survival curve steepness, mean dose, and site size to the fractions of CX cells collected indicates that the sites whose damage results in delay can be enclosed in a sphere of about 1.5μ diameter.

FIGURE 2.21. The Fraction, S , of CX Cells Undelayed by X Rays Delivered in the Presence and Absence of Cycloheximide

The Mitotic Selection Technique M. H. Schneiderman

Last year a comparison of the actual mitotic yield with a calculated mitotic yield was presented. The report²⁰ discussed the difficulties of removing all the cells passing through the selection window and the possibility of improvement by establishing a clone of Chinese hamster ovary (CHO) cells (American Type Culture Collection, ATCC-60, CHO K1a) that was more efficiently selected from mitosis. Further investigation of the factors related to efficient selection has led to a protocol which yields mitotic cell selection values which agree more closely with the expected values.

Cells of the new clone of CHO K1a line are plated between 15 to 20 hr before the start of each experiment. 2.5×10^6 to 3.0×10^6 cells are plated into a 75 cm Falcon plastic flask containing modified McCoy's 5A medium at pH 7.0-7.2 at 37°C. The combination of these conditions, the new clone, and maintenance of the pH and temperature during the entire selection procedure have resulted in consistently good mitotic yields, as shown in Table 2.2.

TABLE 2.2. Theoretical and Experimental Mitotic Yields (CHO K1a)

Selection Time, Minutes	Percent Selected	
	Experimental	Theoretical
220	26.4	24
220	23.4	24
230	30.2	25
220	27.0	24
220	22.7	24
220	19.8	24
220	20.2	24
Average	<u>24.2</u>	<u>24.1</u>

Hydroxyurea: A G_2 Effect

M. H. Schneiderman, D. B. Leeper,* and W. C. Dewey**

The response of cells to treatment with various drugs is important to the evaluation of the effect of radiation

* Thomas Jefferson University, Philadelphia, Pennsylvania.

** Colorado State University, Fort Collins, Colorado.

FIGURE 2.19. The Effect of Low Doses of X Rays in the Presence of Cycloheximide on the Mitotic Yield of Chinese Hamster Ovary Cells

FIGURE 2.20. The Effect of Low Doses of X Rays in the Presence of Cycloheximide on the Mitotic Yields of the Groups of Chinese Hamster Ovary Cells Listed Under Figure 2.16

Figure 2.21a shows the fractions of the CX cells collected, with and without cycloheximide, as functions of the dose. The cycloheximide makes a big difference: without it the fractions form a straight line; with it, a curve. Some understanding of the reasons for these shapes can be found by comparing them with radia-

tion survival curves. The fractions can be analyzed with the same concepts and methods as survival curves because both collection and survival depend on the probabilities of single charged particles or pairs of charged particles producing events in small biochemical entities.

In earlier work,¹⁸ we suggested replotting survival data in the form $-\ln(S)/D$ versus D . Figure 2.21b shows the fractions, now denoted by S , plotted this way. The theory indicates what kinds of curves should fit the data, but here the experimental uncertainties prevent a unique fitting. We present two possibilities in Figure 2.21 that we think bracket the correct interpretation. The dotted lines represent a model in which no delays are initiated by single charged particles, only by pairs of charged particles; in this model, the mean recovery time has to be about 1 min to fit the data. The solid lines represent a model in which both single charged particles and pairs of them initiate delays; the mean recovery time has to be short--a 1/10 min or less, possibly much less. Other possible models incorporate intermediate degrees of single-particle initiated delays and of mean recovery time.

Within this range of possible interpretations, the role of cycloheximide is the same: It mimics a high dose rate. High dose rates minimize recovery by reducing the time between pairs of events. Cycloheximide, on the other hand, must minimize recovery chemically--possibly by interfering with protein synthesis.

The CX cells are much more apt to be delayed than to suffer reproductive death; the parameters for the curves in Figure 2.21 are some hundreds of times larger than the corresponding ones for reproductive survival.

Application of Kellerer's¹⁹ relation between survival curve steepness, mean dose, and site size to the fractions of CX cells collected indicates that the sites whose damage results in delay can be enclosed in a sphere of about 1.5 μ diameter.

FIGURE 2.21. The Fraction, S , of CX Cells Undelayed by X Rays Delivered in the Presence and Absence of Cycloheximide

The Mitotic Selection Technique M. H. Schneiderman

Last year a comparison of the actual mitotic yield with a calculated mitotic yield was presented. The report²⁰ discussed the difficulties of removing all the cells passing through the selection window and the possibility of improvement by establishing a clone of Chinese hamster ovary (CHO) cells (American Type Culture Collection, ATCC-60, CHO K1a) that was more efficiently selected from mitosis. Further investigation of the factors related to efficient selection has led to a protocol which yields mitotic cell selection values which agree more closely with the expected values.

Cells of the new clone of CHO K1a line are plated between 15 to 20 hr before the start of each experiment. 2.5×10^6 to 3.0×10^6 cells are plated into a 75 cm Falcon plastic flask containing modified McCoy's 5A medium at pH 7.0-7.2 at 37°C. The combination of these conditions, the new clone, and maintenance of the pH and temperature during the entire selection procedure have resulted in consistently good mitotic yields, as shown in Table 2.2.

TABLE 2.2. Theoretical and Experimental Mitotic Yields (CHO K1a)

Selection Time, Minutes	Percent Selected	
	Experimental	Theoretical
220	26.4	24
220	23.4	24
230	30.2	25
220	27.0	24
220	22.7	24
220	19.8	24
220	20.2	24
Average	<u>24.2</u>	<u>24.1</u>

Hydroxyurea: A G₂ Effect

M. H. Schneiderman, D. B. Leeper,* and W. C. Dewey**

The response of cells to treatment with various drugs is important to the evaluation of the effect of radiation

* Thomas Jefferson University, Philadelphia, Pennsylvania.

** Colorado State University, Fort Collins, Colorado.

on subpopulations of asynchronously growing cells. Hydroxyurea, a cancer chemotherapy agent, has been reported to kill S-phase cells but to have no effect, particularly on progress in the mitotic cycle, of G₂, M, and G₁-phase cells. A number of investigators report piling up of cells at the G₁-S boundary which tends to confirm this interpretation. The present study was undertaken to determine the effect of hydroxyurea on G₂-phase cells. It was found that, contrary to the previous conclusion, G₂ cells are affected by hydroxyurea.

The progress of G₂ Chinese hamster ovary cells treated with 1.0-mM hydroxyurea was monitored by the mitotic selection procedure. The results are shown in Figure 2.22 along with the mitotic-yield curve for G₂ and M cells that had no treatment other than pulse labeling. (For comparison, the results of x-ray irradiation and of cycloheximide treatment are also shown.) The figure shows that there is a significant change in the kinetics of the G₂ cells following hydroxyurea treatment. The yield is changed from an S-shaped curve into a linearly decreasing one. The hydroxyurea affects the cell progression immediately after it is added. In addition, comparison of the shape of the mitotic-yield curve for the hydroxyurea treatment with the shapes of the curves for x-ray or cycloheximide treatment indicates that probably all of the G₂

cells were affected by the hydroxyurea, none were refractory.

Dose Rate Effects and *Chlamydomonas* L. A. Braby and W. C. Roesch

In previous work,^{21,22,23} we tested a theoretical dose-rate and fractionation model with the green alga, *Chlamydomonas reinhardi*, because its long period of unchanging radiosensitivity made it an ideal test system. We concluded at that time,²¹ on the basis of reports by several groups using a variety of sources of low LET radiation, that the cells were killed by both one-event and two-event processes. Further experimental and theoretical work has led us to change our conclusion: we now believe that only two-event killing takes place. This has clarified our understanding of *C. reinhardi* and paved the way for other experiments.

The first indication that one-event killing might not be significant followed last year's fractionation experiment,²¹ which gave a 35-min mean recovery time. With this value, the theory predicted the low dose-rate survival curve without the assumption of one-event killing that we previously had to make.

Additional evidence was provided by results of a theoretical method we developed to examine other data. The method uses the expression,²¹

FIGURE 2.22. The Affect of Hydroxyurea (O) on the Mitotic Yield of Chinese Hamster Ovary Cells

$$S = \exp \left[-CD - A(\dot{D}\tau)^2 \left\{ D/D(\dot{D}\tau) - 1 + \exp[-D/(\dot{D}\tau)] \right\} \right] \quad (1)$$

for the survival, S , rearranged as follows:

$$- \ln(S)/D^2 - C/D = A(\dot{D}\tau/D) \left[D/(\dot{D}\tau) - 1 + \exp[-D/(\dot{D}\tau)] \right] \quad (2)$$

This expression is a function of dose and dose rate only through their quotient, $D/\dot{D}$ (which equals the irradiation time). Thus, survival data for a variety of doses and dose rates will fall on a single curve if the left-hand side of the equation is plotted versus $D/\dot{D}$. All of the quantities on the left are measured ones except the one-event factor, C ; it has to be found by iteration. Once an appropriate value of C is found, i.e., once the data lie on a single curve, the two-event factor, A , and the mean recovery time, τ , are easily determined.

When tried with our 1973 data on *C. reinhardi*,²² this new method did not give a fit with the one-event factor we had originally deduced; it did fit with the one-event factor equal to zero. We concluded that we had to measure *C. reinhardi*'s survival over a wide range of dose rates both to clear up the nature of its radiation response and to demonstrate the validity of Eq. (2).

C. reinhardi cells, mating type +, were plated on minimal-media agar and covered with silicone oil to prevent drying. They were irradiated with 1.5 MeV, Van de Graaff electrons. The electrons were scattered from a graphite block to reduce the dose rate to allow use of horizontal dishes. Dose rates, from 50 to 3,000 rad/min, were monitored by measuring the current from the scattering block with an electrometer. Doses were determined by integrating the electrometer current with a voltage-to-frequency converter whose output was counted. The monitor was calibrated with thermoluminescent dosimeters (0.015-in. thick LiF, calibrated against a standard radium gamma-ray source). The surviving fraction of cells was determined 3 days after irradiation by

counting both live and dead cells on each dish. Dead cells are as easily observed as live ones, even though they do not divide, because their cell walls can be seen.

The survival curves, Figure 2.23, illustrate a distinct dose rate effect. They are curved and approach a limiting curve at high dose rates; they appear to become straight and less steep at low dose rates. It was deemed impractical to expose the cells longer than 5 hr, therefore, we were unable to show conclusively that they reach the limiting straight line at low dose rates.

Figure 2.24 shows the data plotted as suggested by Eq. (2), with the one-event factor, C , set equal to zero. The solid line represents a fit of the data with a curve having a two-event factor, $A = 2.6 \times 10^{-8} \text{ rad}^{-2}$, and a mean recovery time $\tau = 24 \text{ min}$. We also calculated curves for a number of other values of C , A , and τ .

FIGURE 2.23. Survival Curves for *C. reinhardi* for Different Dose Rates

FIGURE 2.24. The Survival Data for C. reinhardi from Figure 2.23 with the Data for all Dose Rates Transformed to a Common Curve as Described in the Text

(for example, the dashed lines in Figure 2.24 are for mean recovery times 10% higher and lower than 24 min) from which we estimated these values of A and τ are correct to within $\pm 15\%$. The data are not fit well if the value of C assumed is greater than 10^{-5} rad^{-1} . This is a factor of 5 to 10 less than previously used; hence we now hypothesize that it is zero.

We are much more confident of these results than those reported earlier because they were obtained with one group of cells, at one time, in one laboratory and with one radiation source. Also, they are in good agreement with the dose-rate theory and they strengthen our interpretation of the LET effect in C. reinhardi.²² We feel that we can now conduct successful experiments to determine the effect of temperature, oxygen, LET and drugs on the two-event and recovery processes.

Analytical Expressions for Cross Section Data

W. E. Wilson and L. H. Toburen

In order to provide a more accurate representation of the initial proton ionization process in track structure calculations, we are developing a semi-empirical phenomenological model of the cross sections for electron ejection from polyatomic

molecules. The track structure calculations employ the Monte Carlo method which in turn requires, as a source term, probability distributions for emission of electrons as a function of angle and energy. This is the motivation for developing a parameterized analytic expression for the double-differential cross sections. We have concentrated our attention on data for polyatomic molecules, specifically the H_2O molecule since the data became available, because it is these targets which are of most interest in track structure and microdosimetry calculations.

By dividing the experimental cross sections by the Rutherford Cross Section, the major dependence on proton and electron energies is removed. One is left with angular distributions which are relatively simple functions of scattering angle and which vary slowly with ejection energy. The double-differential cross sections (DDCS), $\sigma(\theta, \epsilon)$ are first scaled according to

$$Y(\theta, \epsilon) = \frac{T \left[(\epsilon + I_p)/R \right]^2}{4\pi A_0^2 N_e} \sigma(\theta, \epsilon) \quad (1)$$

where

T = energy of equivalent velocity electron ($E_p/1836$),

I_p = first ionization potential,

$R = 13.6 \text{ eV (Rydberg)}$, A_0 = Bohr radius, N_e = number of weakly bound electrons.²⁴

Representative angular distributions for 0.5 MeV protons on H_2O molecules are shown in Figure 2.25, $Y(\theta, \epsilon)$ for energy losses of $E = \epsilon + I_p$ of 5, 20, and 50 Rydbergs.

The distributions can be generally characterized by a more or less symmetric peak which narrows and shifts toward forward angles with increasing energy loss, and an additional underlying distribution which is an asymmetric but simple function of scattering angle. The peak is associated with the hard, essentially binary, collisions between the proton and an orbital electron; i.e., the so-called binary encounter peak. The asymmetric distribution accounts for the

FIGURE 2.25. Angular Distributions for Electron Ejection from Water Vapor by 0.5 MeV Protons and for Selected Energy Losses of 5, 20, and 50 Rydbergs. The cross sections are scaled according to Eq. (1). The points indicate experimental data, the solid line gives the nonlinear least-squares fit of Eq. (2) to the data.

distant collisions and also the "drag along" electrons which undergo a transition into a continuum state of the atom. These latter electrons are prominent where the velocities of the electron and proton are nearly equal and account for the rapidly increasing cross section as $\cos \theta \rightarrow +1$ in the $E = 20$ curve in Figure 2.25.

We previously made an attempt to represent the angular distributions with a finite series of Legendre polynomials because the resulting coefficients provide useful theoretical

information about the phase shifts in the collision. We found, however, that too many polynomials were required to describe the distribution adequately for later application in track structure codes.²⁵ We have since tried a number of simple empirical formulas using nonlinear least-squares fitting to obtain the coefficients.

An acceptable simulation of the 0.5 MeV proton ejection of electrons from H_2O has been achieved with

$$Y(\theta, \epsilon) = A_1 e^{-Z(\theta)} + A_4 + A_5/K(\theta) \quad (2)$$

where

$$Z(\theta) = [(\cos \theta - A_3)/A_2]^2 \quad (3)$$

and

$$K(\theta) = \sqrt{\frac{Me}{2}} \left[v_p^2 + v_e^2 - 2v_p v_e \cos(\theta) \right]^{1/2} \quad (4)$$

The first term in Eq. (2) is a Gaussian distribution which describes the hard-collision peak, the remaining terms represent the soft collisions and the drag-along electrons. $K(\theta)$ is simply the velocity difference between the outgoing electron and the proton, hence A_5/K becomes large where the electron has a velocity close to that of the proton. This term provides the upturn needed to fit the angular distributions near $\theta = 0^\circ$ and $E \approx 20$ Rydbergs (Figure 2.25).

The solid lines in Figure 2.25 for the three different energy losses are the fit (in a least-squares sense) of Eq. (2) to the real angular distributions. Equation (2) fits the functional form of the data fairly well except perhaps at back angles where it tends to be $\approx 15\%$ too large. This situation probably could be improved by using more terms, i.e., more coefficients. However, experience with the least-squares fitting indicates there is insufficient information in the angular distributions to unambiguously determine more than about five coefficients.

The dependence on energy loss of the set of five coefficients was

obtained by least-squares fitting the angular distributions at energy losses between 2 and 80 Rydbergs and the results are compiled in Figure 2.26. A_1 , which is the amplitude of the Gaussian term, is initially small and monotonically increases to its maximum value at 80 Rydbergs, which is almost exactly the classical maximum energy loss, i.e., $4T$. Above 80 Rydbergs, A_1 is determined merely by inspection inasmuch as there are not enough points in the angular distributions for larger E to determine five coefficients. The width of the Gaussian distribution, A_2 , decreases monotonically between 2 and 80 Rydbergs, approaching a constant value for large energy losses. This behavior reflects the narrowing of the binary-encounter peak with increasing momentum transfer. A_3 is the centroid of the Gaussian term and its functional behavior is to approach asymptotically the cosine of the classical scattering angle with increasing momentum transfer. A_3 is also large for small energy losses because there the rather structureless angular distributions can be represented by a very broad Gaussian which shifts more and more toward forward angles with increasing energy loss (Figure 2.25).

Equations (1-4), with the coefficients shown in Figure 2.26, were used to produce the 3-dimensional plot of the DDCS shown in Figure 2.27. The anomaly near $\cos \theta = +1$ and an electron energy of 300 eV is the representation of the "drag-along" electrons. [Equation (2) has a singularity at $\cos \theta = +1$ and $v_e = v_p$.]

As a test of the phenomenological model, we compare in Figure 2.28 the energy spectrum of electrons ejected at all angles, calculated from the model, with the spectrum actually measured. The single differential cross section times the square of electron energy is plotted against electron energy. For clarity, only every third data point is plotted. One can see that except for the Auger peak at 500 eV, the model simulates the single differential cross section very well. We have not yet included Auger emission in the model; since Auger emission is isotropic and monoenergetic, including its contribution should be a trivial addition. For ejected electrons with energies less than 100 eV, the model tends to be low by up to 10%; and, in the region of 600 eV, the model is, at most, 5% too high.

FIGURE 2.26. Model Coefficients for 0.5 MeV H⁺ on Water Vapor as a Function of Energy Loss, $E = \epsilon + I_p$

FIGURE 2.27. Model Simulation of Double-Differential Ejection Cross Sections for 0.5 MeV Protons on Water Vapor

FIGURE 2.28. Comparison of Single Differential Electron Energy Distributions Calculated from the Model with Experimental Data

Relative probability curves like the one depicted in Figure 2.27 are the desired source terms for the Monte Carlo codes used in track structure calculations. There remains the problem of obtaining the curves for other values of proton energy. We expect to follow the same procedure described above for other energies for which we have data (0.3, 1.0, and 1.5 MeV) and then parameterize the coefficients with respect to incident ion energy.

Microdosimetry of Internal Sources W. C. Roesch

Concern over the possible accidental release of plutonium and transplutonium alpha emitters, especially as particulates, from nuclear power generating plants has raised anew some old questions about the spatial distribution of dose due to internal emitters.^{26,27} We felt that application of microdosimetry to these problems would be fruitful because it deals with the random depositions of energy by independent charged particles and it can be extended to deal with the more complicated depositions near particulates. A new program, Dosimetry of Particulate Sources in the Lung, has been proposed and funded to develop and apply microdosimetric methods to internal emitters. Prior to the start of the new program, much of the basic theory was worked out under the Radiation Dosimetry program. That theory will be described here; the progress toward reducing it to practice will be described in the report for the new program.

The theory developed by Rossi and his colleagues²⁸ needs to be extended to make it apply to internal emitters. Their theory aims at determining the probability density, $f(z)$, of the specific energy, z , in a site. [The specific energy is the energy per unit mass deposited in a small volume called a site. Because of the randomness in energy deposition processes, it may have one of a range of values. The product, $f(z) dz$, is the probability the site received a specific energy in the infinitesimal range dz about z .] This density was all they needed, because they dealt with external photon or neutron radiations, which, within small regions at least, produce the same $f(z)$ for all sites.

Internal sources are often nonuniformly distributed, most notably as particulates; consequently, $f(z)$ is not the same for all sites. Another probability density, $g(z)$, is needed for internal dosimetry--the probability density that a site chosen at random from a selected region has a specific energy in dz at z . In other words, $f(z)$ is the density of a singly stochastic process--the deposition of

energy in a given site; and $g(z)$ is the density of a doubly stochastic process--the selection of a site in the region and the deposition of energy in that site. To put it in a mathematical language: if $f_{dV}(z)$ is the density for any site in an infinitesimal volume element, dV , and $v_{dV}dV$ is the probability of finding a site in the element, then

$$g(z) = \int v_{dV} f_{dV}(z) dV \quad (1)$$

Although the density in specific energy for all sites, $g(z)$, is the goal of microdosimetry of internal sources, the density for individual sites, $f_{dV}(z)$ of Eq. (1), is needed as a step on the way. It is found by compounding the probabilities for deposition of energy in the site by all of the atoms that decay nearby. The mathematical method for doing this, called "convolution," is carried out efficiently with Fourier transforms. The Fourier transform (denoted by $\underline{F}$) of the density was found to be

$$\underline{F} f_{dV}(z) = \exp \left[- [D/(\alpha E)] \int w_{dV} \right. \\ \left. \left[1 - \exp \left[-\alpha \Omega \left(1 - \underline{F} \{ f_{dV'}(z;1) \} \right) \right] \right] dV' \right] \quad (2)$$

Here, $w_{dV'} dV'$ is the probability of finding a particulate in the volume element dV' , α is the mean number of decays in the particulates, E is the energy of the charged particles released in each decay, Ω is the constant of proportionality between α and the mean number of energy depositions in the site, and $f_{dV'}(z;1)$ is the probability density in specific energy for the site when it is known that there has been a single energy deposition event in it due to a decay in a particulate in dV' . An equation for $f_{dV}(z)$ could be obtained from Eq. (1) by taking the inverse Fourier transform (and defining a notation for it), but those familiar with past work in microdosimetry can actually work better with the transform itself.

The absorbed dose, D , that appears in Eq. (2) is the average dose, the dose that would exist if the activity in the region were uniformly dispersed. Internal dosimetry and radiation protection workers often use the average dose because it is the easiest quantity for them to obtain. It is used here to simplify applying microdosimetry in these fields, although its actual function in the theory, in the factor $D/(\alpha E)$, is to give the number of particulates per unit mass.

The single-event density, $f_{dV'}(z;1)$ in Eq. (2), is the basic information needed to apply the theory. It is different for each distance between the site and the decaying atom and, for particulates large enough that emerging charged particles lose an appreciable part of their energy, it also depends on particulate size. A single-event density also appears in the Rossi theory, but there is only one for all sites and the theory assumes that it is measured rather than calculated. Here, on the contrary, measurements are assumed to be too difficult or too numerous to be practicable. The Dosimetry of Particulate Sources in the Lung program includes determination of $f_{dV'}(z;1)$ by Monte Carlo simulation methods.

Experimental Microdosimetry for Alpha Particles

L. A. Braby and W. C. Roesch

The results presented in this discussion are meant to be a specific test of the previously discussed theory of dose distribution from particulate sources in a uniform medium. Distributions of energy density must in general rely on calculations as measurements would be too difficult and too numerous to be practicable. Nevertheless, a few measurements in situations chosen to critically test the assumptions and methods of the calculations can lend confidence to the study or indicate aspects of the theory that need to be strengthened.

In the course of developing computer codes, an experimental check was identified for which we had the necessary apparatus.²⁹ This involves determining the probability density in specific energy due to alpha emitters uniformly and randomly dispersed in a homogenous medium. This was one

of the distributions which represents a limiting case in the calculations. This case can be simulated by a wall-less proportional counter in a large volume of gas containing radon.

A proportional counter in the form of a right cylinder (height equal to diameter) formed by a grid of wires was located inside a large chamber that contained the counting gas. The counter could be operated at gas pressures simulating a site diameter as small as $3 \mu\text{g}/\text{mm}^2$ and yet provide an effective infinite medium for the emitted alpha particles. The counter resolution was 16%, full width at half maximum, for ^{55}Fe x rays. This resolution is typical for counters of this type. Radon was added to the counting gas by placing a radium-painted instrument dial on the floor of the large container and waiting for several days until radon became abundant in the gas.

The solid curve in Figure 2.29 is the probability density in specific energy produced by a point source of ^{241}Am placed five counter diameters from the counter or, in this case, $23 \mu\text{g}/\text{mm}^2$. For comparison, the dashed curve was calculated by the computer program for alpha particles from a point source at a similar distance from a spherical absorbing volume. The agreement is reasonable considering that the calculated density is an approximate one pending completion of Monte Carlo studies and the fact that ^{241}Am has a number of different energy alpha particles and other radiations. Furthermore, the experimental result reflects a cylindrical volume which the calculation is for a spherical site. The conclusion is that the experimental apparatus is essentially operating properly.

The solid curve in Figure 2.30 is the density of specific energies mea-

FIGURE 2.29. The Probability Density in Specific Energy for Alpha Particles from Single Point Sources: (—) Experimental Data for ^{241}Am ; (---) Calculated for Plutonium Alpha Particles Having the Same Stopping Power at the Site

sured with radon in the gas; the dashed curve is the calculated density. They disagree. There are several possible reasons, none of which have we yet had time to examine. For example, the radon or its daughters may be attached to something in the system rather than diffused through the gas. We thought that the daughters, being nongaseous, would settle on the container walls out of range of the counter. If many of them deposited instead on the gridwork of the counter, they could account for the large peak observed at 60 rad. Another possibility is that the difference is partially due to using a right cylinder for the experiment and a sphere for the calculations. Although not conclusive yet, this measurement illustrates the need for experimental work in support of the calculations.

FIGURE 2.30. The Probability Density in Specific Energy for Alpha Particles from Point Sources Uniformly and Randomly Dispersed in a Homogenous Medium: (-) Experimental Data for Radon; (--) Calculated for Plutonium Alpha Particles

• DOSIMETRY OF PARTICULATE SOURCES IN THE LUNG

The purpose of this study is to develop practical methods for calculating microdosimetric distributions for soft tissue and for lung tissue in which plutonium or other alpha-emitting elements are deposited, particularly when deposited as particulates, to aid in correlating and extrapolating radiation effects measured at different levels of exposure and in different species. Computational methods will be developed and tested in the Radiological Physics Section; concurrently, the Radiological Health Section will develop cell and tissue models in which those methods will be applied. The work on this newly funded program has just begun. The first version of a computer program for particulate microdosimetry and some initial results with it are reported here; related theoretical work is reported under the Radiation Dosimetry program.

Calculations for Microdosimetry of Internal Sources W. C. Roesch

Concern over the potential for the release of plutonium or transplutonium alpha emitters, especially as particulates, from nuclear power generating plants has raised anew some old questions about the spatial distribution of dose due to internal emitters. As a result, the Dosimetry of Particulate Sources in the Lung program was started with the aim of applying microdosimetric methods to lung-deposited alpha emitters. The microdosimetry of Rossi and his colleagues was developed for external sources and needed supplementing for application to internal sources. This was done under the Radiation Dosimetry program and is reviewed under that part of this report. Now, the modified theory has to be translated into computer programs to calculate microdosimetric probability densities for a wide variety of internal-emitter deposition patterns.

The computer work has been divided into two parts that can be pursued independently. Work on one part, a Monte Carlo simulation of energy deposition by single alpha particles, has just begun. Progress on the other part, a program for combining the depositions by single alpha particles to give the deposition by internally deposited emitters, is reported here. Later the two parts have to be joined; until then an approximation to what is expected from the Monte Carlo study is being used in the other part.

A first version of the program for combining energy depositions has been completed. In this version, all the emphasis was put on developing accuracy and speed in combining probability densities, because the bulk of the expensive computer time goes into this process. The complications due to inhomogeneity of the lung and to energy loss within large particulates were left to be dealt with later.

Densities are combined by either calculating the first few terms of an infinite series, using the discrete (fast) Fourier transform, or approximating with a normal distribution of the correct mean and variance--depending on the average number of alpha-particle events.

The first version is already adequate for the microdosimetry of modest-size particulates in soft tissue, the liver for example, even though postponing the inclusion of inhomogeneity and particulate size makes it not yet adequate for the lungs. The accompanying figures show an example calculated with the program: Alpha particles from stationary, point plutonium particulates placed at random in a homogeneous medium irradiate sites 4.66μ in diameter. The site size affects the results obtained in microdosimetry; selecting the right size is part of the investigation. The $4.66\text{-}\mu$ diameter was chosen because it gave a D_0 for a track-segment experiment (simulated with the same computer program) of 75 rad, a value typical of experimental results for mammalian cells.

Figure 2.31 shows the probability densities in the specific energy (the energy per unit mass in the site) for single alpha particles emitted at different distances from the center of the site. These are the approximate densities that have been assumed pending the completion of the Monte Carlo studies. They are based on known track-length distributions. These were broadened to simulate straggling and transport of secondary radiations, and a low specific-energy component was added to simulate secondaries from alpha particles that do not themselves pass through the site. This secondary component was estimated from our experimental results with wall-less proportional counters.

Figure 2.32 shows the probability densities in specific energy for particulates that emitted different mean numbers of alpha particles. The mean numbers range from 1 to 10^5 alpha particles, and the number of particulates changes inversely so that the total number of alpha particles remains the same--enough to give an average absorbed dose of 75 rad. The areas beneath the curves range from 0.48 to 0.0022. The missing area

FIGURE 2.31. The Probability Density in Specific Energy for Single Energy-Deposition Events Due to Plutonium Alpha Particles from Point Sources at Different Distances from the Center of a Site 4.6μ in Diameter

FIGURE 2.32. The Probability Density in Specific Energy in Sites 4.6μ in Diameter for an Average Dose of 75 Rad Delivered by Plutonium Alpha Particles from Stationary Particulates, Uniformly and Randomly Dispersed in a Homogeneous Medium, and Having Mean Numbers of Decays Per Particulate Between 1 and 10^5

(the difference from 1.0) equals the probability that no alpha particle or secondary particle deposited energy in a site. The mean specific energy, counting the zero depositions, is 75 rad for each particulate size; however, the means for just the curves,

not counting the zeros, range from 156 to 34,100 rad. In other words, the probability of a site not being hit at all at 75 rad is quite high, but if it is hit, a large energy is deposited within it.

The absorbed dose, 75 rad, is small enough that alpha particles from single particulates are responsible for most of the shape and position of the curves in Figure 2.32. If the dose were increased, each curve would at first keep about the same shape, but the area beneath it would increase; then the curves would begin to shift toward higher specific energies as the effects of more than one particulate become important; finally, at high doses, the curves would shift steadily toward higher specific energies and their shapes tend toward that of a normal distribution.

As an example of how microdosimetric information can be used, consider the estimation of the fraction of cells that survive an irradiation. For alpha particles, this fraction must nearly equal the probability of zero energy deposition: if there is no deposition, the cells are unaffected (barring secondary effects of deposition in neighboring cells); if there is deposition, the specific energy will probably be high enough to be lethal. The probability of zero energy deposition can be shown to be an exponential function, e^{-KD} , of the dose, D . For survival curves, K is

often called the "radiation sensitivity;" it is the inverse of D_0 . Figure 2.33 shows values of K for three different site radii as a function of the number of alpha particles per particulate. The radiation sensitivity, as defined, decreases as the particulates get larger.

FIGURE 2.33. An Estimate of the Radiation Sensitivity, K in the Expression e^{-KD} for the Surviving Fraction, for Irradiation by Plutonium Alpha Particles from Stationary Particulates, Uniformly and Randomly Dispersed in a Homogeneous Medium, as a Function of the Mean Number of Alpha Particles Emitted by the Particulates

REFERENCES

1. W. E. Wilson and L. H. Toburen, "Electron Emission from Proton-Hydrocarbon-Molecule Collisions at 0.3-2.0 MeV," Phys. Rev. A, vol. 11, pp. 1303-1308, 1975.
2. D. J. Lynch, L. H. Toburen and W. E. Wilson, "Electron Emission from Methane, Ammonia, Monomethylamine, and Di-Methylamine by 0.25 to 2.0 MeV proton," J. Chem. Phys., (in press).
3. Y.-K Kim, "Energy Distribution of Secondary Electrons, I. Consistency of Experimental Data," Radiation Research, vol. 61, pp. 21-35, 1975.
4. Y.-K. Kim, Mitio Inokuti and R. P. Saxon, "Slow Electrons Ejected from Rare Gas Atoms by Fast Charged Particles," ANL-8060, Part I, Argonne National Laboratory, pp. 52-54, 1973.
5. J. B. Crooks and M. E. Rudd, "Angular and Energy Distribution of Cross Sections for Electron Production by 50-300 keV Proton Impacts on N₂, O₂, Ne, and Ar," Phys. Rev. A, vol. 3, pp. 1628-1634, 1971.
6. R. C. Der, R. J. Fortner, T. M. Kavanagh and J. M. Khan, "Production of Carbon K X Rays by Heavy Ion Bombardment," Phys. Rev. A, vol. 4, pp. 550-562, 1971.
7. A. Langenberg and J. Van Eck, "Fluorescence Yield of Carbon," Phys. Rev. Lett., vol. 31, pp. 71-73, 1973.
8. J. D. Garcia, R. J. Fortner and T. M. Kavanagh, "Inner-shell Vacancy Production in Ion-Atom Collisions," Rev. Mod. Phys., vol. 45, pp. 111-177, 1973.
9. M. L. West, "Quenching of Benzene Fluorescence in Pulsed Proton Irradiation," J. Phys. Chem., (in press).
10. A. Kupperman, "Diffusion Kinetics in Radiation Chemistry," The Chem. and Biol. Action of Rad., vol. 5, pp. 84-166, 1961.
11. A. Ganguly and J. Magee, "Theory of Radiation Chemistry III," J. Chem. Phys., vol. 25, pp. 129-134, 1956.
12. M. Matheson and L. Dorfman, Pulse Radiolysis, The MIT Press, Cambridge, MA, p. 160, 1969.
13. M. L. West, "Pulsed Radioluminescence," BNWL-1850, PT4, Battelle-Northwest, Richland, WA, pp. 49-52, February 1974.
14. Stopping power for carbon and hydrogen taken from A. Green and L. Peterson, "Energy Loss Functions for Electrons and Proton in Planetary Gases," J. Geophys. Res., vol. 73, pp. 233-241, 1968. Fraction of energy deposited in core taken from W. Brandt and R. Ritchie, "Initial Interaction Mechanisms in the Physical State," in Physical Mechanisms in Radiation Biology, USAEC Technical Information Center, p. 31, 1974.
15. F. Hirayama and S. Lipsky, "Fluorescence of Saturated Hydrocarbons," J. Chem. Phys., vol. 51, pp. 3616-3617, 1969.
16. G. Beck and J. K. Thomas, "Pico-second Observations of Some Ionic and Excited-state Processes in Liquids," J. Phys. Chem., vol. 76, pp. 3856-3863, 1972.
17. M. H. Schneiderman, W. C. Dewey, D. B. Leeper and H. Nagasawa, "Use of the Mitotic Selection Procedure for Cell Cycle Analysis: Comparison Between the X-ray and Cycloheximide G₂ Markers," Exptl. Cell. Res., vol. 74, pp. 430-438, 1972.
18. W. C. Roesch, "Hit Theory: Dose Rate and Energy Density Effects," BNWL-1850-PT4, Battelle-Northwest, Richland, WA, pp. 55-64, February 1974.
19. A. Kellerer, "Analysis of Dose-Effect Relations," Biophysical Aspects of Radiation Quality, pp. 95-114, IAEA, Vienna, 1966.

20. M. H. Schneiderman, "The Mitotic Selection Technique," BNWL-1950-PT4, Battelle-Northwest, Richland, WA, p. 67, February 1974.
21. W. C. Roesch, "Split-Dose Theory," BNWL-1950-PT4, Battelle-Northwest, Richland, WA, pp. 62-65, February 1975.
22. D. S. Nachtwey and L. A. Braby, "LET and Recovery in Chlamydomonas," BNWL-1950-PT4, Battelle-Northwest, Richland, WA, pp. 65-66, February 1975.
23. W. C. Roesch, "Theory of LET Effects," BNWL-1950-PT4, Battelle-Northwest, Richland, WA, pp. 69-71, February 1975.
24. W. E. Wilson and L. H. Toburen, "Electron Emission from Proton-Hydrocarbon-Molecule Collisions at 0.3-2.0 MeV," Phys. Rev. A, vol. 11, no. 4, pp. 1303-1308, April 1975.
25. W. E. Wilson and L. H. Toburen, "Analytical Expressions for Cross Section Data," BNWL-1950-PT4, Battelle-Northwest, Richland, WA, pp. 54-56, February 1975.
26. W. J. Bair, C. R. Richmond and B. W. Wachholz, "A Radiobiological Assessment of the Spatial Distribution of Radiation Dose from Inhaled Plutonium," WASH-1320, U.S. Atomic Energy Commission, Washington, DC, September 1974.
27. "Alpha-Emitting Particles in Lungs," NCRP Report No. 46, National Council on Radiation Protection and Measurements, Washington, DC, August 1975.
28. H. H. Rossi, "Microscopic Energy Distribution in Irradiated Matter," in Radiation Dosimetry, (F. H. Attix, W. C. Roesch, and E. Tochilin, eds.), second edition, vol. I, pp. 43-92, Academic Press, New York, NY, 1968.
29. L. A. Braby, "Microdosimetry," BNWL-1950-PT4, Battelle-Northwest, Richland, WA, pp. 71-72, February 1975.

PUBLICATIONS AND PRESENTATIONS

Publications

Criswell, T. L., L. H. Toburen and W. E. Wilson, "Energy and Angular Distributions of Electrons Ejected from Ar by 5 to 2000 keV Proton Impact," in Abstracts of Papers of the IX International Conference on the Physics of Electronic and Atomic Collisions, Ed. by John S. Risley and R. Geballe (University of Washington Press, Seattle), pp. 749-750, 1975.

Lynch, D. J., L. H. Toburen and W. E. Wilson, "Electron Emission from Methane, Ammonia, Monomethylamine and Dimethylamine by 0.25 to 2.0 MeV Protons," Bul. Am. Phys. Soc., vol. 20, p. 1451, 1975.

Manson, Steven T. and L. H. Toburen, "Energy and Angular Distribution of Electrons Ejected from Ar by 1 MeV Proton Impact Ionization: Theory and Experiment," in Abstracts of Papers of the IX International Conference on the Physics of Electronic and Atomic Collisions, Ed. by John S. Risley and R. Geballe (University of Washington Press, Seattle, WA), pp. 751-752, 1975.

Manson, Steven T., L. H. Toburen, D. H. Madison and N. Stolterfoht, "Energy and Angular Distributions of Electrons Ejected by Fast Protons and Electrons: Theory and Experiments," Phys. Rev. A, vol. 12, pp. 60-79, 1975.

Ratcliffe, C. A., "Programmable Power Supplies Useful for Electrostatic Analyzers," Rev. Sci. Instr., vol. 46, pp. 1586-1587, 1975.

Toburen, L. H. and Steven T. Manson, "Effect of the Cooper Minimum on Charged Particle Ionization," Chem. Phys. Lett., vol. 30, pp. 114-115, 1975.

Toburen, L. H. and W. E. Wilson, "Differential Ionization Cross Sections for 1.3 MeV C^+ Ions on Gas Targets of Hydrogen, Argon and Methane," Bul. Am. Phys. Soc., vol. 20, p. 1451, 1975.

Toburen, L. H. and W. E. Wilson, "Time-of-Flight Measurements of Low-Energy Electron Energy Distributions for Ion-Atom Collisions," Rev. Sci. Instrum., vol. 46, pp. 851-854, 1975.

West, M. L., "Quenching of Benzene Fluorescence in Pulsed Proton Irradiation," J. Phys. Chem., (in press).

Wilson, W. E. and L. H. Toburen, "Electron Emission from Proton-hydrocarbon-molecule Collisions at 0.3-2.0 MeV," Phys. Rev. A, vol. 11, pp. 1303-1308, 1975.

Wilson, W. E., L. H. Toburen and W. A. Glass, "Energy Straggling Parameters from Single Event Spectra," Rad. Res., vol. 63, pp. 387-394, 1975.

Presentations

Lynch, D. J., L. H. Toburen and W. E. Wilson, "Electron Emission from Methane, Ammonia, Monomethylamine and Dimethylamine by 0.25 to 2.0 MeV Protons," Paper presented at the Division of Electron and Atomic Physics (APS) Meeting, Tucson, AZ, December 1975, and paper accepted for publication in the J. Chem. Phys., 1975.

Manson, Steven T. and L. H. Toburen, "Energy and Angular Distribution of Electrons Ejected from Noble Gases by Proton Impact Ionization," Fourth International Conference on Beam Foil Spectroscopy, Gatlinburg, TN, September 1975.

Porter, L. E. and L. H. Toburen, "Distributions in Energy and Angle of Electrons Ejected from Sulfur Hexafluoride by 0.3 to 1.8 MeV Protons," Winter Meeting of the American Physical Society, Anaheim, CA, January 1975.

Toburen, L. H. and W. E. Wilson, "Differential Ionization Cross Sections for 1.3 MeV C^+ Ions on Gas Targets of Hydrogen, Argon and Methane," Paper presented at the Division of Electron and Atomic Physics (APS) Meeting, Tucson, AZ, December 1975.

HEALTH PROTECTION

- **DOSIMETRY OF INTERNAL EMITTERS**

• DOSIMETRY OF INTERNAL EMITTERS

This program is generally aimed at providing dosimetric guidance and support to radiobiology studies at Pacific Northwest Laboratories. In particular, the program includes considerations of dosimetric design of experiments, materials, theoretical and experimental dosimetry of various tissues and eventual relating of dosimetry data to observed biologic effects.

The distribution of radiation dose in the walls of the G.I. tract from ingested, insoluble beta emitters was both calculated and measured. The median lethal ingested ^{106}Ru - ^{106}Rh activity concentration in adult dogs was about 3 mCi/kg (2.75 mCi/kg produced a "delayed" death, $^{180}\text{LD}_{50}$, whereas 3.5 mCi/kg produced an "acute" death, $^{10}\text{LD}_{50}$). The median lethal concentrations in rats for acute death were about 9 mCi/kg ^{106}Ru - ^{106}Rh ; 17 mCi/kg ^{91}Y and 5000 mCi/kg ^{147}Pm . The LD_{50} dose to critical tissue (crypt cells) in the dog gut was about 4000 rads with a 95% confidence interval of 1500-6500 rads. The LD_{50} dose to critical tissue in rats was about 3300 rads with a 95% confidence interval of 2500-4100 rads; values very close to those determined for the dog.

Dose to G.I. Tract from Ingested Insoluble Beta Emitters

F. T. Cross,* G. W. R. Endres,*
F. N. Eichner,* R. A. Libby** and
M. F. Sullivan†

The distribution of dose inside the walls of tissue-equivalent cylinders of various radii-containing material uniformly contaminated with insoluble beta emitters was calculated using Berger's tables of scaled absorbed-dose distribution.¹ The methodology was patterned after

Unnikrishnan et al,² with the dose calculations spanning the energy range 0.225-3.54 MeV β^- max and radii range 0.05-5.00 cm to accommodate the size of the G.I. tract of various animals which might be used for ascertaining the dose associated with acute G.I. lethality. Similar to the findings of Unnikrishnan et al, the dose at a particular depth increased with the internal radius of the cylinder, the amount of increase being more the higher the beta energy. Also, the increase became larger as the depth increased. The values of dose at zero depth were found to be very close to the half dose delivered to the contents, the methodology of ICRP,³ although the Berger derived

* Physics and Instrumentation
Department

** Nuclear Technology Department

† Biology Department

values tended to decrease when the range of the betas became large compared to the radius of the cylinder simulating the segment of the G.I. tract.

The applicability of the calculations to the estimation of the dose to the G.I. tract from ingested insoluble beta emitters is dependent upon subsequent knowledge of the concentration of the radioisotope and its residence time in the various segments of the G.I. tract as well as the radius of the particular segment. The dose to the G.I. tract is described by the following equation:

$$D_{G.I.} = D_{Berg} \frac{ft}{m} \quad (1)$$

where

D_{Berg} = the dose distribution per $\mu\text{Ci hr}^{-1} \text{ g}^{-1}$ for cylinders of various radii corresponding to segments of the G.I. tract,

f = fraction of the ingested activity present in the various segments of the G.I. tract,

t = residence time in hours for the fraction f , and

m = mass in grams of the segment contents.

It is assumed, as in the model proposed by Eve,⁴ that there is a particular residence time during which the various segments of the G.I. tract may be supposed to be filled

with contaminated food and that the distribution of the activity in these segments is uniform.

A summary of the radiobiological data developed in the Pacific Northwest Laboratories animal experiments aimed at producing G.I. death from insoluble beta emitters administered by gavage and by contaminated food is shown in Table 3.1.

Estimation of Lethal Dose to Dog G.I. Tract. TLD-100 rectangular rods (1 x 1 x 6 mm) sealed in plastic were sutured in the G.I. tract wall of dogs to provide an estimate of the dose received by various segments of the G.I. tract as well as to check the accuracy of the dose calculations. A mean and standard deviation of the measured data from eight animals is included in Table 3.2. The spread in some cases is very large and represents not only the errors in the dose measurements but the errors in estimating and correcting for the depths of dosimeter implantation as well as the estimation and correction for the depths to the target cells.

As the majority of deaths (although not all) resulted from damage to the midcolon, it appears reasonable to assign the dose to the mucosal surface to be 8000 rads and the dose to the critical tissue to be 5000 rads with a 95% confidence interval of about 2000-8000 rads for an administered concentration of 3 1/2 mCi/kg ("acute" lethality level). Although death is delayed, it still occurs at the lower concentration of 2.75 mCi/kg resulting in a measured critical tissue dose

TABLE 3.1. Summary of Radiobiological Data

Rat $^{10}\text{LD}_{50}$ (large bowel)	9 mCi/kg (^{106}Ru) 17 mCi/kg (^{91}Y) 5000 mCi/kg (^{147}Pm)
Dog $^{10}\text{LD}_{50}$ (large bowel)	3.5 mCi/kg (^{106}Ru)
Dog $^{180}\text{LD}_{50}$ (large bowel)	2.75-3.0 mCi/kg (^{106}Ru)
Depth to target tissue	0.0750 cm (dog) 0.0175 cm (rat)
Diameter of G.I. tract segment	1.5 cm (dog colon) 0.4-0.5 cm (filled rat colon) ~1.5 cm (filled rat cecum)

TABLE 3.2. Dose Measurements (rads) in Dogs in Various Segments of the G.I. Tract from ^{106}Ru - ^{106}Rh

Location	Target Cell Depth (cm)	Surface Dose for "Acute" Lethality ($^{10}\text{LD}_{50}$)	Critical Tissue Dose for "Acute" Lethality ($^{10}\text{LD}_{50}$)	Critical Tissue Dose for "Delayed" Lethality ($^{180}\text{LD}_{50}$)
Stomach/Pylorus	0.060	700 $\pm$ 400	470 $\pm$ 270	370 $\pm$ 210
Stomach/Curvature	0.060	1100 $\pm$ 360	760 $\pm$ 240	590 $\pm$ 190
Duodenum	0.075	600 $\pm$ 150	380 $\pm$ 95	300 $\pm$ 75
Jejunum	0.125	1100 $\pm$ 670	540 $\pm$ 330	430 $\pm$ 260
Ileum	0.050	2000 $\pm$ 530	1500 $\pm$ 380	1100 $\pm$ 300
Cecum	0.065	3600 $\pm$ 1700	2300 $\pm$ 1100	1800 $\pm$ 890
Upper Colon	0.075	6500 $\pm$ 1500	4100 $\pm$ 970	3200 $\pm$ 760
Midcolon	0.075	8100 $\pm$ 2600	5100 $\pm$ 1600	4000 $\pm$ 1300
Lower Colon	0.075	12,000 $\pm$ 8200	7400 $\pm$ 5200	5800 $\pm$ 4100

of about 4000 rads with a 95% confidence interval of about 1500-6500 rads.

The calculated doses for an acute lethality concentration of 3 1/2 mCi/kg are 8400 and 5200 rads to the surface and critical tissue, respectively. The equivalent doses at a 2.75 mCi/kg concentration calculate to be 6600 and 4000 rads. It can be reasonably concluded, therefore, that the LD_{50} dose in the dog is on the order of 4000 rads to critical tissue in the gut from ingested, insoluble beta emitters with a possible lower limit of about 2000 rads.

Estimation of Lethal Dose to Rat G.I. Tract. Values of ft/m were determined from passage time studies using ^{106}Ru - ^{106}Rh as a tracer and from measurements of the mass of the gut contents. These values for the large bowel of 200 $\pm$ 7 g rats are 1.8 (cecum), 1.7 (upper large intestine, ULI) and 1.3 (lower large intestine, LLI) with an approximate

standard error of 30%. The calculated doses to the large bowel are shown in Table 3.3.

As expected, the dose to the surface mucosa varies greatly; however, the dose to the critical tissues is nearly alike for all three radioisotopes. The mean and standard deviation at the critical depth for these three radioisotopes is:

Cecum	3300 $\pm$ 410 rads
ULI	2400 $\pm$ 730 rads
LLI	1800 $\pm$ 540 rads

The dosimetry suggests that the preponderant damage occurs in the cecum, a fact borne out by the pathology. As a consequence, it is concluded that the LD_{50} dose from ingested, insoluble beta emitters in the rat is on the order of 3300 rads with a range (95% confidence interval) of 2500-4100 rads; values very close to those determined for the dog.

TABLE 3.3. Median Lethal Doses to Rat G.I. Tract

	^{106}Ru - ^{106}Rh			^{91}Y			^{147}Pm		
	Cecum	ULI	LLI	Cecum	ULI	LLI	Cecum	ULI	LLI
Surface Dose (rads)	4200	2000	1500	3700	2600	1900	1.2×10^5	1.1×10^5	8.2×10^4
0.0175 cm Dose (rads)	3600	2000	1500	2900	2000	1500	3500	3300	2400

REFERENCES

1. M. J. Berger, "Distribution of Absorbed Dose Around Point Sources of Electrons and Beta Particles in Water and Other Media," MIRD Pamphlet No. 7, J. Nucl. Med., vol. 12: Supplement No. 5, 1971.
2. K. Unnikrishnan, et al., "Dose Distribution within a Cylindrical Wall Containing a Solution of β -emitters, with Application to the G.I. Tract," Proceedings of IAEA Symposium, Dosimetry Techniques Applied to Agriculture, Industry, Biology and Medicine, Vienna, p. 243, 1973.
3. Recommendations of the International Commission on Radiological Protection, Report of Committee II on Permissible Dose for Internal Radiation, Pergamon Press, 1959.
4. I. S. Eve, "A Review of the Physiology of the Gastrointestinal Tract in Relation to Radiation Doses from Radioactive Materials," Health Physics, vol. 12, p. 131, 1966.

ANALYSIS AND EVALUATION

- **TOXIC SUBSTANCES IN NUCLEAR FUELS**
- **ASSESSMENT OF SOCIAL VALUES IN THERMAL PLANT SITING**
- **CONSIDERATIONS AND METHODOLOGIES FOR STUDIES CONCERNING THE EFFECT OF LOW-LEVEL EXPOSURES TO ENVIRONMENTAL CONTAMINANTS**

• TOXIC SUBSTANCES IN NUCLEAR FUELS

The objective of this program is to characterize toxic materials present in the nuclear fuel cycle and to provide quantitative projections of the controlled accumulation and routine release of these materials. Detailed characterization of the amounts and the chemical, physical, and radiological properties of toxic effluents is required to accurately assess the environmental impact of nuclear fuel cycle facilities. The relative importance of the fuel cycle effluents will be established, and the more important unknown physical and chemical characteristics of effluents will be identified as candidates for further investigations in biomedical and environmental research and development programs designed to refine the environmental impact analysis of nuclear fuel cycles.

Toxic Effluents from UO₂ Fuel Cycle R. M. Fleischman, W. A. Muckerheide, K. J. Schneider and R. L. McKeever

The implementation of nuclear energy systems poses potentially serious questions about the ecological and health effects of routine effluents and the potential release of materials from large accumulations of toxic materials. Measuring the health and ecological effects of pollutants released to the environment requires collecting data over long periods of time. Therefore, it is important that where additional experimental data are required, these be identified as early as possible to give guidance to experimental programs designed to refine the environmental impact analysis.

Assumptions that affect the accuracy of environmental impact analysis, and usually result in conservatism, may be introduced not only in the characterization of the released materials (i.e., source terms) but also in the models that use the source term data to predict environmental transport, human uptake and conse-

quent health effects. The effort to date has emphasized characterization of the effluent source terms and identification of the state of the art in this area. Continuing studies will be directed at identifying other points in the environmental impact analysis where assumptions are made due to the lack of definitive data and assessing their impact on the results of environmental analysis as a key to future investments in experimental programs.

The effort to characterize toxic effluents from the LWR UO₂ cycle has been completed utilizing multidisciplinary expertise from throughout the laboratory. For each fuel cycle component (i.e., mining through waste disposal) effluents have been identified and characterized, to the extent possible, by their radiological, chemical and physical properties. Whereas the radiological and physical form of the effluents are reasonably well known, there is generally a paucity of data available on effluent chemical properties which are, intuitively, key inputs to environmental and biological pathway analysis.

Draft reports of the operating characteristics and effluent characterization efforts for the LWR UO_2 fuel cycle components have been prepared and a final integrated report is in preparation. Continuing studies will be directed at assessing the

importance of the unknown effluent characterization data relative to environmental and biological impact analysis to add perspective to determining where R&D investments are needed.

• ASSESSMENT OF SOCIAL VALUES IN THERMAL PLANT SITING

The objective of the project is to develop a method for combining social values with techno-economic data for better decision making in thermal power plant siting. Quantified technical impact data which is socially weighted will allow for the optimal choice of site/design alternatives. In this phase, a study of community leadership in energy/environment issues was conducted in a town familiar with a (nuclear) thermal power plant. Secondly, a linear composite analysis of the measurement problems associated with combining diverse scales of impacts was completed. Third, a revision of the social values instrument used in a previous phase of the study was initiated. Finally, liaison activities with utilities were undertaken. Quantification of three of the technical impact factors is currently underway.

Social Values in Thermal Plant Siting

J. B. Burnham, J. C. King and
J. A. Hébert

It has been demonstrated in a previous phase of this project that weighting techno-economic data by social values is a feasible undertaking [Contract AT(45-1): 1830, BNWL Report 1787]. In that portion of the study, a model in which quantified technical values of impacts (established by experts) were multiplied by social values (established by representative citizens) was outlined.

A key objective of this phase was to test the feasibility of identifying "representative" community leaders in energy/environment issues. With assistance from the Electric Power Research Institute, Palo Alto, California, the test site of San Clemente, California, was chosen. Southern California Edison, the area utility, was consulted and informed of the project. Two different nomination procedures were investigated. The first procedure asked members of

the business community and residents active in issues of environmental preservation to nominate leaders in the community on energy/environment issues. Those nominated were in turn asked for their nominations until no new names appeared. The same procedure was applied to a random sample of 25 telephone subscribers in the San Clemente area. The first procedure was very efficient at identifying the leadership, but the second was found to be very disappointing and inefficient. Few of the random sample could or would nominate an energy/environment leader while the business/environmentalist sample produced many overlapping nominations.

A second thrust of the project was an analysis of the scaling issues involved in combining diverse quantities such as aesthetics and water quality, especially with reference to social weighting. The weight (importance) of a technical impact was shown to be determined by three factors: 1) the variance of the scale values of the quantified technical

measurement; 2) the covariance of the scale values with those of the other scale values and 3) the social weight assigned. It was shown that great care must be taken that the social values are independent of the variance and covariance information.

Revision of the social values instrument has been initiated to reflect fossil fuel alternatives in addition to nuclear site/design options. The addition of fossil fuel alternatives enhances generalizability of the research and represents a realistic set of options for the short-term.

Liaison activities with utilities will be stepped up, since they would

ultimately be one of the potential users of this research methodology. Preliminary contact with EPRI and Southern California Edison has been made.

Finally, quantification of three of the seven technical factors identified in previous work has been undertaken. Air Quality, Water Quality and Plant/Animal Life factors are in the process of being quantified by experts.

The next phase of the project will see the quantification of the remaining technical factors and full scale test of the methodology in a community facing an energy decision.

- **CONSIDERATIONS AND METHODOLOGIES FOR STUDIES
CONCERNING THE EFFECT OF LOW-LEVEL EXPOSURES
TO ENVIRONMENTAL CONTAMINANTS**

The overall purpose of this project is to assemble and review relevant information, define inherent difficulties of low-level exposure studies, discuss the tractability of these problems, and evaluate and adapt epidemiological and statistical methods for studying them. The primary focus is on studies where ionizing radiation is of concern.

This program was initiated in fiscal year 1976. Our principal concern this year is the analyses of Hanford mortality data, an undertaking in which we not only hope to elicit considerable information on the relationship of radiation exposure to mortality patterns, but also one in which we expect to gain experience and develop methods for dealing with this type of data.

Statistical Considerations of the
Effect of Low-Level Exposures to
Environmental Contaminants

E. S. Gilbert, R. L. Buschbom, and
E. B. Perrin

The problem we are concerned with is how to best evaluate and analyze available data on the effects of low-level exposures to environmental contaminants, particularly ionizing radiation. Our initial goal is a detailed analysis of Hanford mortality data. Our efforts to date have been spent gaining the necessary background to enable us to utilize the available information in as meaningful and informative a manner as possible.

In June of 1975 we completed a draft of "An Evaluation of Milham's Analyses of Hanford Deaths" which generally confirmed Milham's conclusions but was critical of his statistical methods. We have since

completed a final version of our analysis of the Milham data. It was as a result of this document that we were requested by ERDA to undertake an analysis of the full Hanford mortality file.

In September, E. S. Gilbert, R. L. Hooper, and W. J. Bair (from Pacific Northwest Laboratories) and E. B. Perrin (Battelle-Seattle) attended a meeting in Washington, DC, of the Steering Committee for the Hanford and Oak Ridge studies. At this meeting we discussed the results of the Milham analysis, agreed on certain comparable methodologies and generally shared ideas on possible approaches to analyzing this data.

Also as a result of the September meetings, we became aware of a program¹ developed at Harvard, which utilizes cause-age-time-sex-race-specific mortality rates to determine the expected number of deaths in a

study group. We have since obtained this program, adapted it to our system and anticipate that it will prove a very useful tool in analyzing our data.

In mid-November, we obtained three files of information for over 30,000 Hanford employees. These contain birth and occupational information, yearly external exposure data and mortality data. We are currently trying to coordinate these files into a single file suitable for our purposes. This task includes coding the occupational data.

We hope to have some preliminary analyses completed by February 1976. These include a comparison of our data with U.S. vital statistics both for proportional mortality and population based risks. By the end of the fiscal year we should have a detailed proportional mortality analyses completed utilizing dosimetry and occupational data. We should also have completed some comparisons involving a number of variables (such as years at Hanford and radiation exposure) for the total Hanford data base.

In addition to the above, we have spent considerable time acquiring familiarity with other studies (such as those described in the BEIR² and U.N.³ reports) concerning the effects of low-level radiation exposure on man, as well as researching various approaches to analyzing survival data (such as relative life tables,⁴ multiple regression techniques,⁵ and use of the Weibull distribution).⁶

We have taken advantage of the expertise available in several Battelle-Northwest Departments to inform ourselves on the proper interpretation of our data on radiation exposure as well as conditions at Hanford (such as exposures other than radiation) which may be relevant to our analyses.

Although the main thrust of our efforts has been geared to the analysis of a particular data set, a substantial portion of our work has been relevant to the more general problem of assessing and developing techniques for handling low-level exposure data.

REFERENCES

1. R. R. Monson, "Analysis of Relative Survival and Proportional Mortality," Computers and Biomedical Research, vol. 7, pp. 325-332, 1974.
2. "Report of the Advisory Committee on the Biological Effects of Ionizing Radiations," The Effects on Populations of Exposure to Low Levels of Ionizing Radiation, National Academy of Sciences, National Research Council, Washington, DC, 1972.
3. "United Nations Scientific Committee on the Effects of Atomic Radiation," Ionizing Radiation: Levels and Effects, A report of the Committee to the General Assembly, with annexes, United Nations, New York, 1972.
4. F. Ederer, L. M. Axtell, and S. J. Cutler, "The Relative Survival Rate: A Statistical Methodology," Nat. Cancer Institute Monogr., vol. 61, p. 101, 1961.
5. D. R. Cox, "Regression Models and Life Tables" (with discussion), J. R. Statist. Soc. B. vol. 34, pp. 197-220, 1972.
6. R. Peto, and P. Lee, "Weibull Distribution for Continuous-Carcinogenesis Experiments," Biometrics, vol. 29, pp. 457-70, 1973.

PUBLICATIONS AND PRESENTATIONSPublications

Burnham, J. B., W. S. Maynard and G. R. Jones, "Quantified Social and Aesthetic Values in Environmental Decision-Making," Siting of Nuclear Facilities, Vienna: IAEA, 1975.

Hébert, J. A., M. K. Lindell, W. S. Maynard, S. M. Nealey and J. B. Burnham, "Socially Weighted Linear Composites in Environmental Decision Making," Battelle Memorial Institute, Human Affairs Research Centers, 1975. (Topical Report)

Presentation

Fleischman, R. M. and R. C. Liikala, "Isotopic Composition and Radiological Properties of Uranium in Selected Fuel Cycles," BNWL-SA-5375, Battelle-Northwest, Richland, WA, 1975, Presented at ERDA Conference on Occupational Health Experience with Uranium, Arlington, VA, April 28-30, 1975.

REGIONAL STUDIES

- **PACIFIC NORTHWEST REGIONAL ASSESSMENT PROGRAM**

- **PACIFIC NORTHWEST REGIONAL ASSESSMENT PROGRAM**

The Pacific Northwest (comprised of the states of Alaska, Idaho, Montana, Oregon, Washington and Wyoming) can by several measures be regarded as a national "warehouse" of fossil energy resources. This condition coupled with an evolving national policy stressing utilization of fossil fuels in the near term prior to development of more advanced technologies for energy supply, could result in the imposition of major changes in the region's environmental, socioeconomic and possibly health status.

The objective of the Pacific Northwest Regional Assessment Program is to establish and exercise an integrated analytical assessment program for evaluation of these potential changes that may result from various energy development or conservation scenarios. Such scenarios may themselves result from federal development policies and programs, from regionally specific actions by the states and energy industries, or from actions taken by international factors currently importing energy resources into the region.

The majority of energy related actions characteristically have time scales measured in decades--long planning and implementing periods are involved and the life cycle of actions are several tens of years in length. Similar time scales therefore apply to the magnitude of the environmental, health, and socioeconomic consequences. The cumulative effect of the addition of small increments of stress undoubtedly extend this time scale even further.

After consideration of a variety of approaches to integrated assessment at a regional level, Pacific Northwest Laboratories (PNL) has concluded that dynamic simulation techniques provide the best available approach to evaluating the issues pertinent to the Northwest. As a result, the PNW Regional Assessment Program has been structured in a framework involving ten sectors (shown in Figure 5.1). Each of these sectors involve their own submodels that receive information either from outside the model as exogenous inputs or from other sector submodels.

FIGURE 5.1. Analytical Framework of the Pacific Northwest Regional Assessment Program

As shown in Figure 5.1, there are three primary "driving" sectors--economics, energy and demography. The dynamic results from these sectors in turn provide the primary causes of change in "driven" sectors at the right of the figure. These sectors can also generate constraints (e.g., air quality limits) that can constrain developments. At the present time the model is being formulated to run to the year 2025.

In addition to the several sectors of the dynamic simulation model, a final task has as its main objective the identification and analysis of major institutional constraints to energy development in the Pacific Northwest region. In this study institutional is defined as the formal and/or public arrangements at all governmental levels which set the parameters within which energy development-related impacts may occur. A second and concurrent element of this task focuses on the identification and analysis of how such institutional constraints affect community, regional and state planning and impact mitigation in areas affected by energy development.

At the present time, dynamic simulation techniques that have been developed by others and appropriate to the various sectors shown in Figure 5.1 are being closely examined and the best available methods are being adopted and adjusted to meet the needs of the program. Current progress and plans call for completion of working submodels, incorporation of some regional data and functional relationships, and partial integration of these submodels by June 1976.

Initial application of the analytical framework will be to the interlaboratory cooperative program on the National Coal

Assessment. Additional problems for attention include: 1) the implications of phase out of imports of Canadian natural gas, 2) the long-term implications of the Pacific Northwest Hydro-Thermal Program, 3) the effect of the tentatively proposed transcontinental oil pipeline serving the northern tier states from Puget Sound, 4) the significance of possible load management techniques on electrical generation capacity requirements, and 5) the long-term regional consequences of energy centers.

The following pages provide information on the simulation framework on a sector by sector basis.

Energy--Economic Sectors

J. W. Currie, L. E. Erickson,
D. W. Fraley, T. P. Harrington

Energy resources development and conservation activities, energy economics and macroeconomic activity are the primary factors that will influence the energy related future of the Pacific Northwest. These factors (coupled with demographic considerations) will in turn determine the environmental, socioeconomic and health consequences of alternative paths of development.

Energy and economics are inextricably linked together in any effort directed toward providing a base of information and an assessment framework that can be used by decision makers and the public for analyzing candidate energy policies or actions. As a consequence, these two sectors of the overall simulation framework are presented together.

The role of modeling activity in obtaining the desired information is related in a pictorial fashion as shown in Figure 5.2. At the outset, information necessary to the program will be both quantitative and qualitative as well as consisting of scenarios indicating the type of problems which are to be addressed. Certain portions of the data will be utilized by the model to provide forecasts relative to given scenarios. Model results (information) are then combined with other information to provide assessments.

This task begins by describing as accurately as possible the regional

FIGURE 5.2. The Role of the Model to Regional Assessment

energy-economic system and formalizing this description into a computerized model. Initial efforts have dealt with the types of questions to be addressed by the model and, thus, with the identification of the most important descriptions of the energy-economic system that lend themselves to quantification. These descriptors (state variables) are being linked by time-dependent functions to form a conceptual and theoretically consistent economic framework. Empirical functional relationships and coefficient estimates from previous research will form an initial base for the model. We do not believe that a large data gathering effort is warranted until the base model structure with reasonable estimates of data and coefficients has undergone an initial round of sensitivity testing. This analysis will probably reinforce some initial

hypotheses about how the system responds to changes in parameter values. However, it is expected that at this stage, respecification of certain functional relationships and intensive data gathering effort for the initiation of more rigorous statistical analyses may be necessary in order to adequately describe the system behavior.

Role of Energy-Economic Submodel. Figure 5.3 shows the general role of the energy-economic submodel (inside dashed rectangle) to the overall model. This submodel along with the demographic submodel drives the system.

In its present conceptual state the macroeconomic activity submodel (box 1, Figure 5.3) consists of the

FIGURE 5.3. Role of Energy-Economic Submodel to the General Regional Assessment Model

12 basic industries and 7 secondary industries shown in Table 5.1.

An important element of a successful energy model is the integration of energy related sectors with a macroeconomic model (box 1, Figure 5.3) to evaluate the interaction of differing energy prices with economic growth, other prices and other wages in the economy. Large macroeconomic models are usually unsuited for this task, due to their high degree of aggregation and concentration on problems of effective demand. Production technology is usually based on a constant returns-to-scale assumption. While this assumption may be suitable for processing energy for final demand, it cannot answer questions arising from the extractive nature of primary energy production, characterized by increasing marginal costs of production. Thus, in its present conceptual state, the interaction between variables is not captured. However, this aspect of the research has a top priority at the present time.

Also in the conceptual state is the process by which supplies and demands are matched (box 4, Figure 5.3) to generate absolute and/or relative market quantities and prices. Most large scale models have traditionally employed a mathematical optimization technique (e.g., linear programming) to determine equilibrium prices and quantities.^{1,2} Recent modeling efforts at the Stanford Research Institute³ (SRI) and MIT⁴⁻¹⁰ have employed cost minimizing behavior within an iterative framework without resorting to formal optimization techniques. According to SRI* and MIT,⁷ the major reasons for this approach were that the capital intensiveness of energy production and consumption coupled with existing and potential regulation at different levels create dynamic nonlinear system relationships that cannot be adequately handled within a linear programming framework. Thus, for the reasons stated above, it is planned that the market clearing mechanisms in the energy-economic submodel will be patterned after the MIT and SRI efforts.

* Personal Communication with E. G. Cazalet and D. Nesbitt, Stanford Research Institute, Menlo Park, CA.

TABLE 5.1. Tentative List of Industries for Economic Activity Submodel

	SIC Codes	Title
Basic Industries	01-09	Agriculture, Fisheries, and Forestry
	10-14	Mining
	20	Food and Kindred Products
	24	Lumber and Wood Products
	26	Paper and Allied Products
	28	Chemicals
	29	Petroleum and Coal Refining
	32	Stone, Clay, and Glass
	33	Primary Metals
	34-36	Fabricated Metals and Machinery
	37	Transportation Equipment
	21,22,23,25,27	Other Manufacturing
	30,31,33,39	
Secondary Service Industry	15-17	Construction
	40-42,44-47	Transportation
	48,49	Utilities
	50-59	Wholesale and Retail Trade
	60-69	Finance, Insurance, and Real Estate
	70-89	Services
	91-97,43	Government

Although the energy-economic submodel may eventually describe regional and subregional production and consumption processes and provide quantitative information on such variables as prices, quantities, incomes and employment in different sectors, the submodel will only assess the impacts of what economists refer to as "marketable" production and consumption.

The impacts of a class of products called externalities or "extra-market" products will not be assessed by the energy-economic submodel. An example of an extra-market product or externality (negative in this case) would be SO_x emissions from a coal-fired powerplant. The extra-market production will be generated by the

energy-economic submodel; however, physical impacts of these extra-market products will be described by the submodels concerned with air quality, land and water, social and psychological and bioproductivity impacts. These sectors ultimately should provide feedback information on resource quantity and quality to the energy-economic submodel which not only maintains consistency in the dynamic system but provides the capability for assessing the effects of environmental and economic policies over time. These submodels will also provide inputs to the impact evaluation submodel which will ultimately be provided by the Environmental Protection Agency - DBER project on the measurement of social costs of western energy development.

Energy Supply System. A comprehensive list of energy resource would include the following basic fuels: natural gas, coal, wood, crude oil, uranium, geothermal, tidal, wind, solar, hydro, shale oil, liquified gas and methane. Each energy resource may be treated as flowing through a series of stages to its ultimate end use as shown in Figure 5.4. The stages follow those given by Albers and Black¹² and are described below;

1. Exploration: the exploration for new energy resources and expenditures on technological advancement.
2. Extraction: the gathering of the energy resource and its preparation for transport to primary processing.
3. Transport 1: the moving of energy resource from the point of extraction or importation to the point of primary processing or export.
4. Trade 1: the exchange of the energy resources in the form they left the extraction stage from one economic sector to another and/or from one region or subregion to another.
5. Processing: utilization of the energy resources to produce usable products through various production processes.
6. Transport 2: the moving of processed fuel from the point of processing or importation to the point of conversion or export.
7. Trade 2: (also most of box 4, Figure 5.3) the exchange of processed fuels from one economic sector to another and/or from one region to subregion or another.

8. Conversion: the conversion of primary fuels to their consumption form and the technological conversions necessary to accommodate the use of new fuels.
9. Distribution: the distribution of the consumable fuels to their end points of consumption.
10. Consumption: (also box 3, Figure 5.3) the final consumption of consumable energy products by society.

Energy Conversion. The electric conversion submodel is currently based upon a FORTRAN version of Andrew Ford's "Dynamic Model of the United States Electric Utility Industry, 1950-2010."¹² We have elected to base our simulation in FORTRAN because it is the most well-known computer language and it is readily available on most machines. At this point, the conversion of Ford's DYNAMO-based simulation is reasonably complete. Data is being acquired to regionalize the model.

While Ford's model is for the electric conversion stage, it exemplifies the characteristics of the conversion stage of the energy network (box 8, Figure 5.4). The model may be used to show the behavior of such factors as electricity price, generating reserve margin, and the average rate of return on investments in response to other state variables.

Figure 5.5 provides an overview of the key factors utilized and determined by the model; as is shown, the demand for electricity is determined exogenously to this sector of the energy-economic model. This demand in turn drives electricity production

FIGURE 5.4. The Stages of Energy Flow in the Energy-Economic Model

and the construction of new generation capacity. As can be seen from Figure 5.5, cost calculations are made comparing fossil and nuclear generation, augmented with expected regulatory constraints, and new construction is determined by a capacity addition computation.

Present activities are directed toward adding the linkages to the model which are characterized by Figure 5.4 for the overall energy network. In addition, the first round of data necessary to embody the Northwest region's unique characteristics is being collected to drive the model. Specific changes may be expected in order to effect a regionalization of the model.

Present plans call for the energy-economic submodels to be formalized early in 1976 using best estimates for parameters, coefficients and functional relationships. Integration within the general systems model is planned for completion in June 1976.

Energy Consumption. The last two stages of the energy network are the

distribution and final consumption stages. The distribution stage distributes the consumable energy products to their points of consumption, or to the regional boundary if the energy is exported. For the purposes of the energy economics submodel, these end use sectors (box 10, Figure 5.4 and box 3, Figure 5.3) are, at the present time, as listed below:

1. Residential
2. Industrial
3. Commercial and Government
4. Transportation
5. Agricultural
6. Other

The residential consumption sector, for example, focuses on the household where the primary causal factor will be the number of households, which is provided by the demographic submodel. The household energy demand is also dependent upon a number of other demographic, economic and technical factors. These factors include the type of home, the base appliances and secondary appliances, the annual energy use per appliance, the household income level

FIGURE 5.5. The Electrical Conversion Stage of the Energy-Economic Model

and the price of fuel. The calculation of annual energy consumption could then involve multiplying the total number of household units having each type of appliance by the annual energy use per appliance parameter as in the Wisconsin Energy Model.¹³ Here, four specific factors are accounted for by the specification of their attributes:

1. Type of home (urban single family, urban apartment and rural).
2. Ownership fractions for three "base appliances" (space heating, water heating and central air conditioning).
3. Ownership fractions for 14 "secondary appliances" (e.g., refrigerator, freezer, dishwasher, stove, TV, etc.).
4. Annual energy use per appliance.

The manner in which the above factors may fit into the overall structure of the residential energy consumption submodel is shown in Figure 5.6. For a given year, the

FIGURE 5.6. Example Residential Energy Consumption Submodel Structure

simulation could begin with the determination of a demand for new homes, including that resulting from the creation of new families and from the demolition of old homes. The next step in the sequence would be the calculation of the distribution among the three housing types. These could be determined by either past trends or may be specified by the model user according to the scenario under study. Forecasts for appliance ownership would then be combined with an appliance use model incorporating the influence of future fuel availability, prices of fuels and household incomes.

Demographic Sector J. J. Jacobsen

Energy related development activities in the relatively sparsely populated regions of the Pacific Northwest may result in significant demographic changes particularly at a subregional scale. Although demographic forecasts are routinely prepared by state and Federal agencies, few, if any, of these take into account possible subregional perturbations that may result from major energy development activities. In addition, individual demographic changes may present more significant stresses to environmental quality and the socioeconomic conditions of a region than the existence of the energy facility per se. As a result, the Pacific Northwest simulation model both drives and, in turn, is driven by the demographic sector.

The demographic sector of the model simulates the interactions among the population, the birthrate, the death rate, the net migration rate and the jobs available in the region in response to exogenous inputs and inputs from other model sectors. In turn, the demographic sector will provide other model sectors with information about the number of households, the size of households, the labor force and the age structure of the population. This information will be used to determine energy demand, water usage, land requirements, demand for services and health impacts. The major elements of the demographic sector and some of

their interactions are shown in a causal diagram* in Figure 5.7.

As shown by the four arrows pointing toward population, the population in a region can be determined by four factors: 1) the birthrate, 2) the death rate, 3) the labor force, and 4) the labor force participation rate.** The birth rate and death rate are initially assumed exogenous to the model and constant. As the model is refined, these assumptions may be modified. Information about the number of jobs available in the region from the economic sector is compared with the regional labor force to give the regional unemployment rate. The regional unemployment rate partially determines the regional net migration rate and the labor force participation rate.

For example, if the regional unemployment rate is relatively low, it will be easier for people to find employment and people will enter the

labor market; as a result, the regional labor force participation will be relatively high. Conversely, if the unemployment rate is high, people seeking employment will become discouraged and leave the labor market resulting in a lower labor force participation rate.

The number of people in the regional labor force (employed persons and people seeking employment) is increased or decreased by the labor force migration rate. The net migration rate is, in turn, assumed to be a function of the regional wage rate and the regional unemployment rate relative to the national wage rate--the lower the regional unemployment and the higher the regional wages, the greater number of people migrating into the region.

Figure 5.7 also shows the feedback loop mechanism that works to keep the regional labor force in balance with the total regional jobs. This loop is emphasized in Figure 5.7. If, for example, the regional unemployment rate should drop, people would tend to migrate into the region increasing the regional labor force, which in turn will bring the unemployment rate back up to a "normal or equilibrium level."

This structure is closely related to the demographic sector of a model developed by Battelle-Columbus Laboratories for the Susquehanna River Basin Utility Group.¹⁴

As presently envisioned, the population will be categorized by age and by household size. This structure is shown in Figure 5.8. It can be viewed as a two-dimensional array, the axis being the age of the head of the household and the household size (number of people in the household). While the exact number of categories along each dimension has not been determined, initially there will be six household size classifications (single thru four or more children) and six age classifications (0 to 18, 19 to 24, 25 to 29, 30 to 44, 45 to 64, and 65+). The movement of the population within this structure will be determined by age specific migration rates, age specific birthrates and age specific death rates.

This structure is adapted from work presently underway by the

* Causal Diagrams: The structural assumptions that make up a dynamic model may be expressed by a causal diagram. In a causal diagram, system information interactions are shown by arrows leading from each element to all other variables that might be influenced by changes in that element. The polarity of each causal influence is indicated by a + or - sign at the head of the arrow. A positive polarity means that an increase in the first element will cause an increase in the second (and a decrease will cause a decrease). A negative polarity signifies that an increase in the first element will produce a decrease in the second (and a decrease will produce an increase). Causal diagrams are rough sketches of the interacting feedback loops in the model. They do not contain enough information to permit a complete understanding of the possible modes of behavior or to analyze the model on a computer, but they do convey the general pattern of major system interactions.

** The labor force participation rate equals the ratio of the people employed or seeking employment to the total number of people able to work.

FIGURE 5.7. Interactions Within the Demographic Sector

FIGURE 5.8. Structure of the Population Within the Model

Willamette Simulation Unit at Oregon State University.¹⁵

At the present time the demographic sector model is being coded and is scheduled to be debugged using trial data from a particular region by February 1, 1976. Data requirements will then be finalized and data collected during February 1976. The sector will be linked with other model sectors during March-June 1976. A report documenting the model will be available by June.

Air Resources Sector

D. S. Renne and D. L. Elliott

The objective of the air quality sector of the Regional Assessment Program is to provide estimates of the air quality, and the information necessary to assess the effects on terrestrial environments resulting from the deposition and washout of airborne pollutants, in the Pacific Northwest due to the development of the region's energy resources. A thorough assessment of these impacts requires detailed knowledge of the strength and geographic distribution of present and future energy related emissions throughout the region, including those of the energy facilities themselves as well as secondary emissions (e.g., automotive emission) resulting from the community and transportation growth that will inevitably occur as resources are developed, and a thorough knowledge of the atmospheric transport and diffusion properties throughout the region.

The energy related pollutant emissions will be obtained as part of the output of the energy/economic sectors of the regional model. Emissions factors obtained from the literature (e.g., reference 16 and 17) will be incorporated into this model to convert the amount of fuel burned or converted in various categories (such as

electrical generation or coal conversion, transportation and space heating) and the amount of particulates released by process industries and agricultural activities into total emissions of sulfur and nitrogen oxides, particulates, trace elements, carbon monoxide and hydrocarbons. The emission inventory can be divided into two basic categories: 1) primary or point source emissions due to large fuel burning or conversion facilities such as power plants and gasification plants, and 2) secondary or area source emissions due to space heating, transportation and related industries.

A regional scale transport and diffusion model which accounts for the spatial and temporal variations in the wind field^{18,19} is being used to estimate ground level air quality conditions of present and future energy development activities. This program has the capability of calculating short-term maximum as well as long-term average pollutant concentrations. This model interpolates the mean winds between 100 and 1000 m above the ground measured every 12 hr by the National Weather Service at approximately 34 sites in the Pacific Northwest (Figure 5.9). Trajectories of pollutant transport are then determined from these wind fields. Standard diffusion relations are used to estimate plume dispersion. Removal processes such as dry deposition and

precipitation scavenging, and chemical transformations are incorporated into the model.

For use in the Pacific Northwest Region Assessment model, calculations will be made using typical annual meteorological data to develop indices of the sensitivity of a grid of receptor points to composite emissions occurring throughout the region. This matrix of sensitivity indices can then be coupled to the other sectors of the regional assessment model to calculate air quality and deposition processes for exogenously specified scenarios. The rate and cumulative amount of surface deposited pollutants will be made available to the bioproductivity sector for computation of effects over time.

To date, a preliminary investigation of transport has been made for six large existing or proposed coal-fired power plants listed in Table 3.2. Figure 5.10 shows the plume trajectories for these plants for a 4-day period in April 1974 computed

FIGURE 5.9. Pacific Northwest Meteorological Stations Utilized in the Interpolated Wind Fields. Additional stations off the grid are used for interpolating winds near the grid boundaries.

FIGURE 5.10. Plume Trajectories for Six Existing and Proposed Coal-fired Power Plants. Trajectory calculations begin at 1600 PST on April 1, 1974.

TABLE 5.2. Coal-Fired Power Plants for Which Plume Trajectories were Computed

Name	Location	Megawatts
Centralia	SW Washington	1400 Existing
Boardman	NE Oregon	500+ Proposed
Pioneer	SW Idaho	1000 Proposed
Jim Bridger	SW Wyoming	2000 Existing
Wheatland	SE Wyoming	1500 Proposed
Colstrip	SE Montana	2000 Proposed

from the trajectory model. The existing wind data is influenced by large topographic features such as the Rocky Mountains, and additional analytic methods are being considered to incorporate the effect of smaller topographic features such as the Columbia Basin and Willamette Valley.

Preliminary computations of ground level concentration, deposition, and scavenging of sulfur dioxide and sulfate from several large coal-fired plants in the Pacific Northwest have also been made. Precipitation scavenging is being computed using mean monthly precipitation data extrapolated to a fine-mesh grid. Ultimately the model will be run with 1 year of data to obtain estimates of annual average ground level pollutant concentrations and surface deposition on a grid with 2000 "receptor points" spaced approximately 35 km apart. The emphasis for the remainder of FY 76 will be on annual average sulfur dioxide and sulfate concentrations associated with existing and planned coal-fired power plants and coal conversion facilities in the Pacific Northwest. This effort will include the development of a coal-related emission inventory for the region.

Land and Water Resource Sectors M. W. Stradley and S. J. Shupe

Future regional developments relating to energy extraction, conversion, transmission and consumption are expected to impact the land and water resources of the region. First order or direct land and water related impacts which can result from such developments are typically con-

sidered in environmental impact statements which are filed for individual projects in accordance with state and Federal statutes. While these statements provide important insights into local impacts they often do not consider the long-term systematic impacts which may arise in a particular region or subregion as a result of sequential and seemingly unrelated developments. The systems approach taken in the Pacific Northwest Regional Assessment Program is directed toward these types of phenomena. Furthermore, through interaction with other sectors such as the demographic, social/psychological, economic and energy, the feedbacks from first order land and water effects can be used to investigate second order effects which are not always obvious when traditional "mechanistic" types of impact analyses are employed.

During fiscal year 1976, efforts in the land and water resources sectors are being directed toward the Powder River area of eastern Montana and Wyoming. Geographically this area will include all or part of seven counties (Powder River, Rosebud, and Bighorn in Montana; and Campbell, Converse, Sheridan, and Johnson in Wyoming). This area has been selected for initial study due to the extensive coal-related developments which have been proposed for that area and also in anticipation of the needs of the National Coal Assessment Program being conducted jointly by the ERDA national laboratories.

Current work includes the development of land use and water resources simulation submodel sectors. The land use submodel will be designed to simulate changes in land use patterns and will be driven by demands from the economic (including agriculture), energy and demographic sectors. The proposed model, shown in terms of a causal diagram (see footnote to Demographic Sector) in Figure 5.11, is based on a previous land use model developed at Oregon State University.²⁰ As seen in the figure; economic, demographic and energy demands generated in other portions of the model operate on the land use submodel to cause shifts in land use patterns primarily from agricultural land to industrial and residential land. These shifts in turn affect the price of the land (both agricultural and residential)

FIGURE 5.11. Land Use Sector Causal Diagram

and agricultural production (computed as a function of agricultural capital and land and bioproductivity). Provisions for simulating land speculation and for directing inputs into the water resources sector are also built into the submodel. Initial model development emphasizes the land use pattern dynamics, with the economic factors being addressed in future efforts coordinated between the economics and land use sectors.

The water resources sector submodel is shown in Figure 5.12. As in the land use submodel, various demands from industrial, agricultural and domestic sources compete for available water as well as influence water quality for a given stream or aquifer. Initially the simulation model is being constructed to consider surface water in the main tributaries of the region. Later plans call for the integration of shallow ground water with subsequent provisions for consideration of deep ground water such as the Madison aquifer which underlies the Northern Great Plains States.

It is not presently planned to model competition for available water as a dynamic phenomenon. Rather available water for domestic, industrial and agricultural use will be allocated in a hierarchical sequence specified exogenously as part of a given scenario. Shortages of available water or adverse changes in water quality* will be monitored as part of the model output and used to identify potential problems in the water resources area which could either preclude or inhibit proposed energy related developments under a given scenario. In this sense the water resources submodel will be less interactive than the land use model and serve as a "flag" in the system. The output from the water resources submodel will be used to modify and evaluate scenarios specified external to the model.

* Water quality parameters monitored by the submodel will be dependent on the water pollution loads associated with primary energy development and secondary effects under given scenarios.

FIGURE 5.12. Water Resources Sector Causal Diagram

The models just described will be generic so that their application of other potential impact areas will be possible.

Social Sector M. E. Olsen

Two different models are being constructed to represent the social sector of the regional dynamic simulation model. The first termed "Energy Use Reduction Model," depicts the likely social consequences of rising prices for (or restrictions on) gasoline, heating fuel and electricity. The second, termed "Project Impact Model," depicts the probable social impacts of large-scale energy development projects (such as nuclear power plants or surface coal mining) on the surrounding area. The main reasons for differentiating these two models are that: (1) the more immediate effects of the two situations are rather different, even though some of their long-term social effects may be similar; and (2) data on the effects of reduced energy use can be aggregated from local areas to the entire region, whereas the effects

of any particular development project are largely restricted to the immediately surrounding area and cannot be aggregated to the entire region.

In the Energy Use Reduction Model, the major immediate consequences of rising energy prices and/or governmental restrictions on energy consumption are the following:

- rising cost of driving an automobile per mile
- shift in population distribution from suburbs into central cities
- movement of population from metropolitan areas to small towns and rural areas
- decline in industrial and business jobs
- price inflation.

Some likely secondary consequences of the above conditions are these:

- increase in multiple family dwellings

- decrease in housing size per person
- declining automobile utilization
- decreased purchase and use of household appliances
- lowered median family income.

In the Project Impact Model, the major immediate impacts of any large-scale development project include the following:

- creation of new jobs in the area, both associated with the project and through secondary economic growth
- population growth in the area
- changes in the socioeconomic class and/or racial composition of the area
- need for additional housing in the area
- need for additional public facilities and services of all kinds in the area.

A number of additional factors will affect a community's ability to absorb a large population influx and provide the housing and services they need, including:

- duration of the project
- current amount of unoccupied housing and under-utilized public facilities and services
- local government funding source
- availability of federal or state governmental financial assistance
- availability of employable service personnel.

As an outgrowth of these immediate and secondary effects of energy use reduction or development projects, an affected community may experience many other subsequent social impacts. Since we cannot at this time predict the precise nature of these possible long-term social impacts, we are constructing four different sets of social indicators to measure the quality of life in a local community

(which could then be aggregated to the regional level). These indicators are applicable to both the Energy Use Reduction and the Project Impact models, and could also be utilized when performing other kinds of social impact assessment. The four sets of community quality of life indicators, with their major subdivisions (but not their actual empirical measures), are listed below:

1. Community resources

- Total wealth
- Knowledge personnel
- Local government

2. Services and facilities

- Housing
- Education
- Medical care
- Public health
- Public protection
- Transportation
- Legal services
- Public welfare
- Social services
- Culture and recreation

3. Social problems

- Crime
- Juvenile delinquency
- Alcohol and drug abuse
- Mental and physical illness
- Suicide
- Family disruption
- Social isolation
- Poverty
- Educational problems
- Unemployment
- Inadequate housing
- Minority discrimination

4. Collective responses

- Participation in and actions by:

civic associations
interest associations
neighborhood associations
ad hoc citizen groups

- Organized political participation
- Development or expansion of governmental programs
- Community and regional planning
- Community conflict and collective violence

At the present time, efforts are being focused on: 1) developing an empirical measure for each separate factor in the social sector, 2) examining the existing literature to identify the presence and direction of probable linkages among these various factors, and 3) determining the crucial information flows between the social and other sectors. These three tasks are all interrelated, and hence are being conducted simultaneously. The present time schedule calls for their completion by the end of January 1976. After that, the remaining tasks will be to: 4) locate or collect the data necessary to determine the strengths of the linkages among the various factors, 5) continually revise the model in light of these empirical findings, and 6) express the model in dynamic equations. These tasks will be conducted during February-June 1976.

Psychological Sector D. H. Summers

A wide range of studies have shown that several different aspects of individual behavior are affected by certain types of changes in an individual's milieu. For that reason, energy developments which--directly or indirectly--produce changes in the immediate human milieu may also have substantial effects on behavior. Unfortunately, findings concerning the impacts of social, economic and physical environmental changes have not been systematically integrated, nor has a framework been developed which would make it possible to predict the psychological consequences of such changes.

This phase in the psychological sector of the Regional Assessment Program has focused on the identification of sources in the literature dealing with the impact of variables in the social, economic and physical environment on psychological variables such as productivity, satisfaction, levels of stress and anxiety, and attitudes towards the environment and energy-related issues, etc. Most of the literature identified to date focuses on the psychological changes produced by changes in noise levels, overcrowding, density, physical, and social arrangements in the neighborhood and community and air pollution.

On the basis of this literature review variables have been identified in each of the other model sectors which may cause or influence the types of psychological impacts associated with energy development (see Figure 5.13). The identification of these input variables is tentative and it is recognized that lack of further appropriate data may necessitate the elimination of some of these original variables.

The next step in this sector of the Regional Assessment Program will be to identify the type of relationship that exists between the input variables (from other sectors in this model) and psychological impacts, such as satisfaction and stress. Additionally, an attempt will be made to identify interrelationships among input variables which can affect the type of psychological impact. This step will be based on what is available from prior research efforts documented in the literature.

The third step, which will be intrasector, will be to identify relationships between psychological impacts and individual actions, using whatever data are available. The severity of the impact will presumably dictate the type of individual response or action, and individual actions will in turn affect the collective (community and government) response. The link between individual action and community response is also the link between the psychological sector and the other sectors of the model, most specifically the sociological sector.

Bioproductivity Sector R. H. Sauer

The bioproduction submodel simulates the production of agricultural products such as grains, forage, and meat. Agricultural production accounts for a significant proportion of income in the Pacific Northwest. The siting and installation of energy related facilities in the Pacific Northwest can be expected to directly or indirectly affect agricultural production through changes in land use, regional economics and distribution of by-products of the energy facility, such as SO_x or salt from cooling towers. The bioproduction

FIGURE 5.13. Psychological Sector Relationships

submodel thus has an intermediate position in the total model as it responds to changes in agricultural capacity and needs, and in turn provides information to the energy and economic submodels, the focal points of the total model.

Any region, whether it is the whole Pacific Northwest or a county in one of the states, cannot be expected to have a uniform environment. Annual temperatures and rainfall, for example, will vary geographically. To incorporate spacial heterogeneity, the region addressed by the model will be divided into environmental areas with uniform characteristics. For example, 5% of the region or 1000 acres could have 15 in. annual precipitation, a mean growing season temperature of 20°C, no added nitrogenous fertilizers and no toxic soil minerals. These characteristics can be static and remain the same for the duration of a model run, or can be dynamic and respond to changes in the model. Some degree of change can be expected as construction and operation of an energy facility affect supplies and demands for agricultural production.

Within each environmental area there will be a provision for more than one agricultural product. For example, in the area described above, wheat may grow on 70% of the area with beef production taking up the remainder. The total production for the region would be the sum of all beef production or wheat production in all environmental areas.

The production of each agricultural product will be estimated from relationships of the form

$$Y_i = A_{ij} (a_i X_{1j} + b_i X_{2j} \dots), \quad (1)$$

where Y is the production of product i in environmental area j ; A is the area (e.g., acres) in area j devoted to product i , a , b , . . . are coefficients for product i associated with the environmental parameters X_1 , X_2 , . . . in environmental area j . The total production of product i will be the sum of all i over j . These relationships are based on annual estimates of environment parameters and thus the model has a time step of one year.

If more detail and a mechanistic approach are needed for bioproduction in the regional model, the Ecosystems Level Model (ELM), developed by the Grasslands Biome to simulate the total grasslands ecosystem, can be used. However, the large size of ELM and the 2-day time step make it presently incompatible with the already large planned regional model with a 1-year time step. Much of the intricate detail generated in ELM would not be used in the regional model.

The bioproduction submodel will require information from the other sectors to operate. The quantity of land devoted to each product in each environmental area will be needed from the land use submodel. Similarly, the quantity of soil toxins as estimated to be deposited by the air resources submodel can be employed to calculate changes over time in bioproductivity. These values should be dynamic as they will change with changing land and product values, and with urbanization.

The size and characteristics of the environmental areas will be provided with the bioproduction model. These values too should be dynamic as changes in availability of irrigation water (from the water resources sector) can make large changes in the agricultural production of an area.

The output of the bioproduction submodel will be the total production for each agricultural product in the simulated region. This information can be used by the economics submodel, for example, as input for supply and demand functions, and by the energy submodel to determine the quantity of nitrogenous fertilizer derived from natural gas or the quantity of diesel fuel needed for cultivation.

Institutional Constraints to Energy Development

M. Curry and M. Greene

The ultimate goal of this task is the analysis of institutional constraints at all governmental levels which: 1) affect energy development, 2) can be used to mitigate the occurrence of adverse impacts and 3) can be used to encourage the occurrence of positive impacts.

The first phase of this task, now nearing completion, involves identification and preliminary analysis of federal level institutional constraints for the various categories of impacts. This process has identified federal powers, agencies and legislation that may affect the nature and extent of energy development-related impacts.

This is not a large-scale effort since substantial impact-specific information exists among other staff responsible for each sector, and such specific constraints (e.g., air quality standards) will be programmed into each sector's submodel. Rather, this phase attempts to provide an overview of the relevant federal institutional constraints and analyzes the planning functions of the identified agencies and legislation to determine how these functions may help integrate adverse impacts and encourage positive impacts resulting from energy development. This phase will also analyze the specific constraints, regulations and laws to examine how such constraints interact and influence each other. The combination of such constraints may work synergistically to produce a result which is not simply the sum of the parts and this possible relationship should be identified and analyzed. This should be particularly useful in defining and analyzing constraints which affect the social, psychological, demographic and possible economic and energy sectors of the model since there may be a few formal institutional constraints focusing directly on these sectors.

The second phase of this task is to identify and analyze institutional constraints at the state and regional levels. It is anticipated that during this analysis the emphasis will be more on the planning legislation, tools and agencies available at the state and regional levels because most environmental guidelines and institutional constraints which affect specific impact sectors are found at the federal level.

Special Assessments

W. H. Swift, and G. L. Wilfert

In addition to the requirement to develop a dynamic simulation framework

for the Pacific Northwest Regional Assessment Program, there are frequent needs to provide timely contributions to interlaboratory cooperative efforts (such as the National Coal Assessment) and to provide ERDA with near-term assessment information on a regionally specific basis. Further, there is a strong requirement to establish and maintain working relationships with regional and state agencies concerned with energy planning in the Pacific Northwest. This latter aspect is important in defining issues, developing common data bases and assuring that the program is responsive to regional as well as national needs. A third aspect involves the development of regional data basis compatible with the needs of the dynamic simulation program and the development of interlaboratory data base sharing and transfers.

For example, an understanding of the current "energy balance" of the states (including the relative dependencies on several energy supply forms) is essential as insight for determining the significance of possible or probable future developments. Although such balance studies are static and not predictive of either developments or effects, they do provide reference points-of-departure.

Preliminary energy balances for the Northwest states are shown in Figures 5.14 through 5.19 for calendar year 1974. These balances are derived from primary data published by the Federal Power Commission,²¹ the Edison Electric Institute,²² the U.S. Department of Transportation,²³ and the U.S. Bureau of Mines.²⁴ Although caution must be applied in using a single year's data as indicative of the present regional status, the balances do suggest a very significant dependence of the contiguous states on the imports of Canadian natural gas, a factor further illustrated in Figure 5.20 and Table 5.3. Since Canadian policies may result in major curtailment of exports in the early 1980's, the significance of this loss of basic energy supply is important in assessing what energy substitutions might possibly occur and what their impacts might be on the environment and regional economics. Quite obviously significant changes in energy balances may well occur in the near future.

In another study, the currently announced^{25,26,27} additions to electrical generating capacity in the contiguous northwest states was analyzed and is reported in Figure 5.21 and 5.22.

PUBLICATIONS AND PRESENTATIONS

PUBLICATIONS AND PRESENTATIONS

Publications

Burnham, J. B., W. S. Maynard and G. R. Jones, "Quantified Social and Aesthetic Values in Environmental Decision-Making," Siting of Nuclear Facilities, Vienna: IAEA, 1975.

Crecelius, E. A., "Analysis of Total Iodine in Milk by X-Ray Fluorescence and Iodide Electrode," Anal. Chem., vol. 47, pp. 2034-2035, 1975.

Crecelius, E. A., "The Geochemical Cycle of Arsenic in Lake Washington and Its Relation to Other Elements," Limnol. Oceanogr., vol. 20, pp. 441-451, 1975.

Crecelius, E. A., M. H. Bothner and R. Carpenter, "The Geochemistries of Arsenic, Antimony, Mercury and Related Elements in Sediments of Puget Sound, Washington," Environ. Sci. Tech., vol. 9, pp. 325-333, 1975.

Criswell, T. L., L. H. Toburen and W. E. Wilson, "Energy and Angular Distributions of Electrons Ejected from Ar by 5 to 2000 keV Proton Impact," in Abstracts of Papers of the IX International Conference on the Physics of Electronic and Atomic Collisions, Ed. by John S. Risley and R. Geballe (University of Washington Press, Seattle), pp. 749-750, 1975.

Hébert, J. A., M. K. Lindell, W. S. Maynard, S. M. Nealey and J. B. Burnham, "Socially Weighted Linear Composites in Environmental Decision Making," Battelle Memorial Institute, Human Affairs Research Centers, 1975. (Topical Report)

Lynch, D. J., L. H. Toburen and W. E. Wilson, "Electron Emission from Methane, Ammonia, Monomethylamine and Dimethylamine by 0.25 to 2.0 MeV Protons," Bul. Am. Phys. Soc., vol. 20, p. 1451, 1975.

Mangelson, N. F., G. M. Allison, J. J. Christensen, D. J. Eatough, M. W. Hill, R. M. Izatt and K. K. Nielson, "Rapid Multi-Element Analysis of Biological Samples Using an Energy Dispersive X-Ray Fluorescence Method," in Trace Element Metabolism in Animals - II, Ed., W. G. Hoekstra, J. W. Suttie, H. E. Ganther and W. Mertz, Univ. Park Press, pp. 439-441, 1974.

Mangelson, N. F., K. K. Nielson, M. W. Hill, D. J. Eatough and L. D. Hansen, "A Potpourri of Proton Induced X-Ray Analyses," Third Conference on Application of Small Accelerators, Ed., J. L. Duggan and I. L. Morgan, North Texas State Univ., 1974.

Manson, Steven T. and L. H. Toburen, "Energy and Angular Distribution of Electrons Ejected from Ar by 1 MeV Proton Impact Ionization: Theory and Experiment," in Abstracts of Papers of the IX International Conference on the Physics of Electronic and Atomic Collisions, Ed. by John S. Risley and R. Geballe (University of Washington Press, Seattle, WA), pp. 751-752, 1975.

Manson, Steven T., L. H. Toburen, D. H. Madison and N. Stolterfoht, "Energy and Angular Distributions of Electrons Ejected by Fast Protons and Electrons: Theory and Experiments," Phys. Rev. A, vol. 12, pp. 60-79, 1975.

Nielson, K. K., M. W. Hill, and N. F. Mangelson, "Calibration and Correction Methods for Quantitative Proton-Induced X-Ray Emission Analysis of Autopsy Tissues," Advances in X-Ray Analysis, vol. 19 (in press).

Rancitelli, L. A., R. W. Perkins, W. D. Felix, and N. A. Wogman, "Solar Flare and Lunar Surface Process Characterization at the Apollo 17 Site," Proc. of the Fifth Lunar Conference, Geochim. Cosmochim. Acta, vol. 2, pp. 2185-2203, 1974.

Ratcliffe, C. A., "Programmable Power Supplies Useful for Electrostatic Analyzers," Rev. Sci. Instr., vol. 46, pp. 1586-1587, 1975.

Toburen, L. H. and Steven T. Manson, "Effect of the Cooper Minimum on Charged Particle Ionization," Chem. Phys. Lett., vol. 30, pp. 114-115, 1975.

Toburen, L. H. and W. E. Wilson, "Differential Ionization Cross Sections for 1.3 MeV C⁺ Ions on Gas Targets of Hydrogen, Argon and Methane," Bul. Am. Phys. Soc., vol. 20, p. 1451, 1975.

Toburen, L. H. and W. E. Wilson, "Time-of-Flight Measurements of Low-Energy Electron Energy Distributions for Ion-Atom Collisions," Rev. Sci. Instrum., vol. 46, pp. 851-854, 1975.

West, M. L., "Quenching of Benzene Fluorescence in Pulsed Proton Irradiation," J. Phys. Chem., (in press).

Wilson, W. E. and L. H. Toburen, "Electron Emission from Proton-hydrocarbon-molecule Collisions at 0.3-2.0 MeV," Phys. Rev. A, vol. 11, pp. 1303-1308, 1975.

Wilson, W. E., L. H. Toburen and W. A. Glass, "Energy Straggling Parameters from Single Event Spectra," Rad. Res., vol. 63, pp. 387-394, 1975.

Wogman, N. A., "Comparison of Ge(Li) and Anti-Compton Systems for Measurements of Environmental Samples," IEEE Transactions on Nuclear Science, 1973 Symposium, vol. NS-21, no. 1, BNWL-SA-4876, February 1974.

Presentations

Carpenter, R. and E. A. Crecelius, "Geochemical Cycles and Budgets of Arsenic in Puget Sound and Lake Washington," NSF-RANN Trace Contaminants Abstracts, 2nd. Annual Conference Issue, August 1974.

Cooper, J. A., H. L. Nielson, N. A. Wogman, and R. W. Perkins, "Feasibility Study of In Situ Sediment Analysis by X-Ray Fluorescence," Nucl. Tech., vol. 26, pp. 224-231, June 1975.

Crecelius, E. A., "Chemical Changes in Arsenic Following Ingestion by Man," Presented at the 15th Hanford Life Sci. Symp., September 29-October 1, Sponsored by ERDA (To be published in ERDA Symposium Series), 1975.

Crecelius, E. A., "The Geochemical Cycle of Arsenic in Lake Washington, Seattle," Presented at the 37th Annual Meeting of the American Society of Limnology and Oceanography, June 23-28, Seattle, WA, 1974.

Crecelius, E. A. and R. Carpenter, "Arsenic Distribution in Waters and Sediment of the Puget Sound Region," In First Annual National Science Fond., Trace Contaminants Conf., Oak Ridge Nat'l. Lab., pp. 615-625, August 1973, 1974.

Crecelius, E. A., C. J. Johnson and G. C. Hofer, "Contamination of Soils Near a Copper Smelter by Arsenic, Antimony and Lead," Water, Air and Soil Pollution, vol. 3, pp. 337-342, 1975.

Crecelius, E. A., D. E. Robertson and L. A. Rancitelli, "The Development and Characterization of Marine Standard Reference Materials (SRM's) for Chemical Speciation Determinations," Presented at the Workshop on SRM's for Marine Science, Catalina Island, Oct. 8-9, Sponsored by National Bureau of Standards, 1975.

Fleischman, R. M. and R. C. Liikala, "Isotopic Composition and Radiological Properties of Uranium in Selected Fuel Cycles," BNWL-SA-5375, Battelle-Northwest, Richland, WA, 1975, Presented at ERDA Conference on Occupational Health Experience with Uranium, Arlington, VA, April 28-30, 1975.

Hill, M. W., D. J. Eatough, L. D. Hansen, K. K. Nielson and N. F. Mangelson, "Aerosol Within a Copper Smelter Analyzed by Proton Induced X-Ray Fluorescence and Calorimetry," Presented at International Nuclear and Atomic Activation Analysis Conference and 19th Annual Meeting on Analytical Chemistry in Nuclear Technology, Gatlinburg, TN, Oct. 14-16, 1975.

Kosorok, J. R., "A Handler for the Versatec Serial Printer Interface," BNWL-B-449, September 1975.

Kosorok, J. R. (speaker) and C. A. Ratcliffe, "Memory Data Storage Via DECtape," DECUS Fall Symposium, San Diego, CA, November 5-8, 1974.

Lynch, D. J., L. H. Toburen and W. E. Wilson, "Electron Emission from Methane, Ammonia, Monomethylamine and Dimethylamine by 0.25 to 2.0 MeV Protons," Paper presented at the Division of Electron and Atomic Physics (APS) Meeting, Tucson, AZ, December 1975, and paper accepted for publication in the J. Chem. Phys., 1975.

Manson, Steven T. and L. H. Toburen, "Energy and Angular Distribution of Electrons Ejected from Noble Gases by Proton Impact Ionization," Fourth International Conference on Beam Foil Spectroscopy, Gatlinburg, TN, September 1975.

Porter, L. E. and L. H. Toburen, "Distributions in Energy and Angle of Electrons Ejected from Sulfur Hexafluoride by 0.3 to 1.8 MeV Protons," Winter Meeting of the American Physical Society, Anaheim, CA, January 1975.

Rancitelli, L. A., J. S. Fruchter, W. D. Felix, R. W. Perkins, and N. A. Wogman, "Cosmogenic Isotope Production in Apollo Deep Core Samples," to be published in Proc. of the Sixth Lunar Conference, Geochim. Cosmochim. Acta., 1975.

Rieck, H. G., Jr., J. R. Kosorok, R. W. Perkins, and N. A. Wogman, "In-Situ X-Ray Fluorescence System Design," BNWL-B-394, Battelle-Northwest, Richland, WA, April 6, 1975.

Toburen, L. H. and W. E. Wilson, "Differential Ionization Cross Sections for 1.3 MeV C^+ Ions on Gas Targets of Hydrogen, Argon and Methane," Paper presented at the Division of Electron and Atomic Physics (APS) Meeting, Tucson, AZ, December 1975.

Wogman, N. A., "Current Methods for Laboratory Analysis of Environmental Levels of Radioactivity," Presented to IEEE-Radionuclide in the Environment Measurement and Monitoring, Accepted for Publication in IEEE Proceedings, to be published June 1976.

Wogman, N. A., H. G. Rieck, Jr., and J. R. Kosorok, "In Situ Analysis of Sedimentary Pollutants by X-Ray Fluorescence," Presented to A.C.S. Mini Symposium, Walla Walla, WA, November 1974, Nucl. Inst. & Meth., vol. 128, pp. 561-568, BNWL-SA-5204, 1975.

ORGANIZATION CHARTS

Pacific Northwest Laboratories

Tommy W. Ambrose, Director

William A. Watts, Assistant to the Director

Program Support

James J. Fuquay, Assistant Director

Research

Douglas E. Olesen, Director

Programs

Jerry W. Finnigan, Director

Finance & Administration

Kurt K. Leaser, Controller and Director

Human Resources

Richard J. Mayer, Facilitator
and Director

Legal & Contract Services

Sam J. Farmer, General Counsel
and Director

Operations & Services

William D. Richmond, Director

Atmospheric Sciences Department
Charles L. Simpson, Mgr.
Charles E. Elderkin, Assoc. Mgr.

Metallurgy Development Department
Robert W. Stewart, Mgr.

Biology Department
William R. Wiley, Mgr.
James F. Park, Assoc. Mgr.

Nuclear Technology Department
Neal E. Carter, Mgr.

Chemical Technology Department
Richard E. Nightingale, Mgr.

Physics & Instrumentation Dept.
A. Judd Haverfield, Mgr.

Ecosystems Department
Burton E. Vaughan, Mgr.
William L. Templeton, Assoc. Mgr.

Radiological Sciences Dept.
Julian M. Nielsen, Mgr.
Richard W. Perkins, Assoc. Mgr.

Engineering Technology Department
Les T. Pedersen, Mgr.
Ben M. Johnson, Assoc. Mgr.

Systems Department
John C. Fox, Mgr.

Fuels & Materials Department
Robert P. Marshall, Mgr.

Water & Land Resources Dept.
Dennis B. Cearlock, Mgr.

Energy Mission
Loren C. Schmid

Biomedical & Environmental Program
William J. Bair, Mgr.

Exxon Nuclear Program
James E. Minor, Mgr.

Fusion Technology Program
Loren C. Schmid, Acting Mgr.

Nuclear Waste Technology Program
Allison M. Platt, Mgr.

RADIOLOGICAL SCIENCES DEPARTMENT
(7HA0)

January 1, 1976

JM (Julian) Nielsen, Manager
FL (Louise) Barton, Secretary

RW (Dick) Perkins, Assoc. Manager
°°°EF (Liz) Leonard, Secretary

#WH (Bill) Collins, Financial Specialist

#GM (Gary) Garnant, Editor/Writer °°°EF (Ermel) Briggs, Quality Assurance Specialist

EI (Earl) Wandling, Specialist,
Building Management

Analytical and Nuclear Research
(7HA5)

Technical Analysis
(7HA6)

Earth and Planetary Chemistry
(7HA7)

Chemical Methods and Kinetics
(7HA8)

NE (Nate) Ballou, Manager
GJ (Jean) Clark, Secretary
*JE (Jane) Peery, Clerk

LJ ("Rip") Kirby, Manager
AC (Art) Case
VW (Bill) Thomas, Jr.

LA (Lou) Rancitelli, Manager
ES (Betty) Getchell, Secretary

WD (Dale) Felix, Manager
°°°EF (Liz) Leonard, Sec.

++LJ (Larry) Alquist
FP (Fred) Brauer
JE (Jon) Fager
RW (Ron) Gales
RL (Dick) Gordon
JH (Jim) Kaye
JM (Jim) Kelley
CR (Bob) Lagergren
WA (Bill) Mitzlaff
PL (Paul) Reeder
DM (Dean) Robertson
JJ (Jim) Stoffels
RS (Bob) Strebin, Jr.
H (Harvey) Tenny

KM (Kris) McFadden
JD (Jerry) Forsythe
DT (Dottie) Harless
BW (Bruce) Killand
PA (Pat) Wodarz

++KH (Keith) Abel
EA (Eric) Crecelius
JS (Jon) Fruchter
+SR (Sammy) Garcia
JR (John) Kosorok
JC (JC) Lau
JD (Don) Ludwick
+++RL (Rick) McKeever
HG (George) Rieck, Jr.
DE (Dave) Robertson
WB (Wyatt) Silker
TM (Terry) Tanner
ME (Mary) Turner
JA (Jim) Young

RL (Ron) Brodzinski
BA (Bruce) Bushaw
JA (Jim) Campbell
PO (Pete) Jackson
JC (John) Kutt
JC (Jim) Langford
HL (Howard) Nielson
KK (Kirk) Nielson
MR (Mike) Petersen
CW (Chuck) Thomas
WC (Wally) Weimer
NA (Ned) Wogman

RM (Rush) Campbell
DR (Don) Edwards
JG (Grant) Pratt

NL (Lois) Abbey
VC (Velma) Beard
LA (Lee) Bond
GG (Gwen) Brodaczynski
DR (Don) Ellis
RM (Rose Ann) Graves
MS (Mike) Rapids
VB (Vi) Saling
LW (Lena) Schock
SC (Shirley) Simpson

LL (Lavern) Baker
°°°EF (Ermel) Briggs
***WL (Will) Butcher
BG (Chris) Christensen
DA (Dave) Cochran
CL (Chuck) Nelson
JH (Jim) Reeves
RW (Ron) Sanders
MR (Monty) Smith

LEGEND: *Part-time employee, included in total.
 **Temporary employee, included only once.
 ***Hourly employee, included in total.
 °°°Dual assignment, included only once.
 +S&E Program, not included in total.
 ++NORCUS appointment, not included in total.
 #Assigned from another organization, not included in total.
 ++Offsite, Marine Research Lab., Sequim, Washington
 +++Offsite, Colstrip, Montana

Total Number of Exempt = 47
Total Number of Non-Exempt = 32

TOTAL = 79

PHYSICS AND INSTRUMENTATION DEPARTMENT
(7H90)

January 1, 1976

AJ (Judd) Haverfield, Manager
SL (Sharon) Gillis, Secretary

ER (Ernie) Job, Bldg. Mgr., PSL

#EH (Ed) O'Claire, Financial Specialist

°°°DA (Dave) Baker, Quality Assurance Specialist

#JL (Jan) Simmons, Editor/Writer
#LO (Lonnice) Foley, Editor/Writer

HL (Hugo) Libby, Staff Scientist
JT (Jim) Russell, Staff Scientist

Surface Science (7H90)	Optics, Lasers & Holography (7H91)	Nondestructive Testing (7H92)	Radiological Physics (7H93)	Radiological Health Research (7H94)
OK (Otto) Harling, Technical Leader BJ (Barbara) Cole, Secretary	WJ (Bill) Coleman, Manager MJ (Marj) Garrett, Secretary	GJ (Jerry) Posakony, Manager DH (Doreen) Waldkoetter, Secretary	WA (Bill) Glass, Manager JA (Judy) Ewing, Secretary	IC (Iral) Nelson, Manager LC (Linda) Stacy, Secretary
LS (Larry) Price DL (Dave) Styris MT (Tom) Thomas	RA (Ron) Dilbeck JS (John) Hartman BP (Percy) Hildebrand DE (Dave) Hufferd LJ (Leslie) Kirihara LD (Lee) Philipp FR (Fred) Reich KL (Ken) Swinth RA (Ray) Walker	FL (Larry) Becker JC (Jim) Crowe TJ (Tom) Davis PH (Phil) Hutton JR (Jim) Skorpik	LA (Les) Braby ##TL (Tom) Criswell DR (Don) Kalkwarf JA (John) Miller CA (Charlie) Ratcliffe WC (Bill) Roesch LH (Larry) Toburen ML (Martin) West WE (Walt) Wilson	°°°DA (Dave) Baker LW (Larry) Brackenbush FT (Fred) Cross GWR (Bill) Endres LG (Leo) Faust GR (Glenn) Hoenes +TB (Tom) Powers DB (Dill) Shiplier JK (Joe) Soldat
Space Sciences (7H95)	Math & Physics Research (7H96)	RE (Bob) Bowey JF (Jack) Dawson ***NE (Norm) Dixon JL (Jim) Eick SJ (Sid) Foreman JC (Joe) Harris GH (George) Lyon CB (Chuck) Perry PL (Paul) Tomeraasen	JE (Jim) Choate °°R (Rudy) Rodriguez C (Chuck) Veverka	RM (Joyce) Bernard FN (Fred) Eichner DL (Dan) Haggard
RA (Bob) Stokes, Manager VA (Virda) Tarr, Secretary	EV (Vey) Allen LL (Larry) Kopf NG (Neil) Nedrow GF (Gerry) Sisco **DE (Dave) Suffield			
JL (Jerry) Berndt KC (Karl) Davis PA (Phil) Ekstrom EW (Ed) Kleckner ##JJ (Joe) Michalsky MN (Mary) Newby RW (Roger) Owen RH (Ron) Severtsen LL (LeRoi) Smith ##JB (John) Swedlund	WR (Bill) McSpadden, Manager *JC (Joanne) Perry, Secretary *PA (Peggy) Snyder, Tech. Typist			
***FI (Polly) Carver **SM (Steve) Gill JR (John) Schmelzer	BH (Bernie) Duane AG (Alan) Gibbs DL (Del) Lessor HB (Harold) Liemohn CA (Clarence) Oster RL (Dick) Richardson JR (Jim) Sheff JW (Joe) Upton			

LEGEND: °°Trainee, included in 7C00 total.
°°°Dual assignment, included only once.
*Part-time employee, included in total.
**Temporary employee, included in total.
***Hourly employee, included in total.
+S&E Program, not included in total.
#Assigned from another organization, not included in total.
##BI Fellow, not included in total.

Total Number of Exempt = 60
Total Number of Non-Exempt = 32
TOTAL = 92

SYSTEMS DEPARTMENT
(7HMO)

January 1, 1976

JC (John) Fox, Manager
LA (Nadine) McJilton, Secretary

#°°°DL (Doug) Gale, Financial Specialist
#°°°DL (Doug) Gale, Quality Assurance Specialist

#JL (Judy) Harris, Editor/Writer
#DE (David) Pittman, Editor/Writer

WH Swift, Regional Studies Project

<u>Economic Analysis</u> (7HMI)	<u>Computer Technology</u> (7HM3)	<u>Planning and Assessment</u> (7HM4)	<u>Information Systems Technology</u> (7HM5)	<u>Statistics</u> (7HM6)
DE (Duane) Deonigi, Manager GS (Gynetta) Patterson, Secretary LF (Leda) Brown, Clerk *CM (Candy) Eliason, Clerk TL (Terrye) Whitaker, Clerk RW (Bob) McKee, Tech. Ldr., Program Evaluations JR (John) Young, Tech. Ldr., Environmental Impact Studies CH (Clarence) Bloomster LL (Lavelle) Clark M (Marvin) Clement BW (Bruce) Cone JW (William) Currie LE (Lee) Erickson TJ (Thomas) Foley CA (Corey) Knutsen TR (Tim) Kula KA (Karen) McGinnis SA (Ananda) Rao WA (Bill) Reardon ***KE (Keith) Yandon RG (Bob) Rau	MD (Marv) Erickson, Manager ES (Edythe) Clark, Secretary GE (Garth) Driver, Tech. Ldr., Operations +AJ (Art) Boland KM (Ken) Busness MR (Myron) Compton VL (Vern) Crow LH (Larry) Gerhardstein PJ (Pete) Hof RE (Bob) Mahan JO (Jim) Schroeder TA (Tom) Seim PC (Phil) Stover SL (Shirley) Bradymire DN (Doris) Gibson ***RD (Rachel) Langevin EW (Gene) Wallace	JB (John) Burnham, Manager CB (Chris) Ford, Secretary PL (Paul) Hendrickson JJ (Jay) Jacobsen JC (Jeffery) King JW (James) Litchfield CL (Craig) McDonald ***MS (Marge) O'Connell GL (Glen) Wilfert	PJ (Paul) Dionne, Manager CW (Cindy) Womack, Secretary LE (Larry) Addison ***LM (Lynn) Burke RA (Robert) Burnett SD (Steven) Burns BM (Billy) Cole RL (Roger) Engel DW (David) Fraley TP (Tim) Harrington ER (Dick) Hill HD (Harlan) Huber KB (Kent) Stewart WV (Warren) DeMier, Tech. Ldr., Application Programming RS (Rosalie) Argo BB (Barbara) Vinson JC (John) Bower ***RF (Richard) Hart AE (Anne) Medford	RL (Richard) Hooper, Manager *ML (Marsha) Blixt, Secretary *ML (Mary Lou) Lomon, Clerk SM (Sharon) Popp, Clerk JA (Jeffery) Buchanan *ES (Ethel) Gilbert JW (James) Johnston EL (Elbert) Kelley, Jr. *JA (Judy) Mahaffey *DD (Donna) Matsumoto JA (James) Merrill WL (Wesley) Nicholson AR (Anthony) Olsen DL (Donald) Stevens MA (Michael) Wincek PZ (Pauline) Aldridge <u>Biometrics Center</u> +JL (Jessie) Burns RL (Ray) Buschbom RO (Richard) Gilbert VM (Maitland) Lee JR (John) Tadlock PE (Paul) Tucker CR (Charles) Watson ***KA (Kim) Bainard SL (Sarah) Owzarski JC (Jacqueline) Wallace

LEGEND: °°°Dual Assignment, included only once.
*Part-time employee, included in total.
***Hourly employee, included in total.
+S&E Program, not included in total.
#Assigned from another organization, not included in total.

Total Number of Exempt = 70
Total Number of Non-Exempt = 22

TOTAL = 92

OTHER BNW CONTRIBUTORS*CHEMICAL TECHNOLOGY DEPARTMENT

Applied Chemistry - H. H. Vantuyl
T. J. Whitaker

NUCLEAR TECHNOLOGY DEPARTMENT

Engineering Systems Analysis -
R. C. Liikala

BATTELLE

HUMAN AFFAIRS RESEARCH CENTERS

Special Projects Section - S. M. Nealey

J. A. Hebert

OFFSITE CONTRIBUTORS

D. L. Lynch - Columbia Basin College
S. T. Manson - Georgia State University
L. E. Porter - University of Montana
H. Paretzke - Institut fur Strahlenschutz, Neuherberg, Germany
W. C. Dewey - Colorado State University
D. B. Leeper - Thomas Jefferson University

* The names listed under each section are participants in the Physical and Analytical Sciences ERDA Programs; other section members are not indicated.

DISTRIBUTION

<u>No. of Copies</u>		<u>No. of Copies</u>	
<u>OFFSITE</u>		2	<u>Office of Nuclear Regulatory Research</u> U.S. Nuclear Regulatory Commission Washington, DC 20555 A. B. Joseph F. G. Lowman
1	<u>ERDA Chicago Patent Group</u> 9800 South Cass Avenue Argonne, IL 60439 A. A. Churm	1	<u>Office of Standards Development</u> U.S. Nuclear Regulatory Commission Washington, DC 20555 E. E. Held
18	<u>Division of Biomedical and Environmental Research</u> U.S. Energy Research and Development Administration Washington, DC 20545 N. F. Barr W. G. Belter R. P. Blaunstein H. D. Bruner W. W. Burr R. L. Butenhoff D. W. Cole, Jr. R. D. Cooper C. W. Edington W. O. Forster J. S. Kirby-Smith J. L. Liverman J. E. Miller C. L. Osterberg J. Swinebroad H. R. Wasson W. H. Weyzen R. W. Wood	163	<u>ERDA Technical Information Center</u>
		2	<u>Argonne National Laboratory</u> 9700 South Cass Avenue Argonne, IL 60439 P. F. Gustafson J. Sedlet
2	<u>ERDA Division of Environmental Control Technology</u> Washington, DC 20545 H. L. Hollister W. E. Mott	2	<u>Atomic Energy of Canada Limited</u> Chalk River, Ontario C. A. Mawson I. Ophel
1	<u>ERDA Division of Production and Material Management</u> Washington, DC 20545 W. L. Lennemann	1	<u>Atomic Energy Establishment Trombay</u> Bombay 73, India A. K. Ganguly
1	<u>ERDA Division of Reactor Development and Technology</u> Environmental and Sanitary Engineering Branch Washington, DC 20545 Chief, Environmental and Sanitary Engineering Branch	1	<u>Atomic Energy Research Establishment</u> Harwell, Berks, England Librarian
		1	<u>Australian AEC</u> Post Office Coogee New South Wales, Australia A. W. R. Wilson
		2	<u>Brookhaven National Laboratory Research Library, Reference Section</u> Information Division Upton, Long Island, NY 111973 L. P. Hatch G. M. Woodwell

Distri-2

<u>No. of Copies</u>		<u>No. of Copies</u>	
3	<u>Centre d'Etudes Nucleaires de Saclay</u> P.O. Box 2, Saclay Fig-sur-yvette (S & O) France A. Barbreau F. Duhamel P. Slizewicz	1	<u>E. I. Du pont De Nemours & Co., Wilmington</u> ATT: Document Custodian Polymer Intermediate Dept. U.S. ERDA Wilmington, DE 19898 V. R. Thayer
2	<u>Comision Nacional de Energia Atomica</u> Buenos Aires, Argentina D. J. Beninson E. Vander Elst	1	<u>ENEA (OECD) Health & Safety Office</u> 38, Blvd. Suchet Paris XVI, France E. Wallauschek
5	<u>Commissariat a l'Energie Atomique</u> Centre D'Etudes Nucleaires de Cadarache BP n°, 13-115 St Paul les Durance, France M. Rzekiecki	1	<u>Eurochemic Library</u> Mol, Belgium Librarian
1	<u>Commissariat a l'Energie Atomique</u> Centre d'Etudes Nucleaires Fontenay-aux-Roxes, BP n°, 6-9226 Fontenay-Aux- Roses, France A. M. Menoux	1	<u>Institute of Geological Sciences</u> Exhibition Road South Kensington London, SW-7 2DE United Kingdom Dr. Stevenson Buchan Chief Scientific Officer Deputy Director
1	<u>Commonwealth Scientific and Industrial Research Organization</u> Aspendal, Victoria, Australia B. B. Hicks	1	<u>Geological Survey Water Resources Div. (Int)</u> U.S. Department of the Interior U.S. Geological Survey Water Resources Division Washington, DC 20242 M. King Hubbert
4	<u>E. I. Du pont De Nemours & Co., Aiken</u> Savannah River Laboratory Technical Information Service Rm. 773A Aiken, SC 29801 A. R. Boulogne K. McMurdo B. C. Rusche W. B. Scott	1	<u>George Washington University</u> 2029 G. NW Washington, DC 20006 C. R. Naeser
		1	<u>Geotechnical Corporation</u> Box 28277 Dallas, TX 75228 W. B. Heroy
		1	<u>Gessellschaft Kernforschung mbH</u> Karlsruhe 5, West Germany H. Krause

Distri-3

No. of
Copies

No. of
Copies

1 Health & Safety Laboratory
U.S. ERDA
376 Hudson St.
New York, NY 10014

J. Harley

1 International Atomic Energy
Agency
1010 Vienna, Kaerntnerring 11,
P.O. Box 590
Austria

Director, Division of
Nuclear Safety and
Environmental Protection

5 Lawrence Radiation Laboratory
University of California
Lawrence Livermore Lab
Technical Information Dept.,
L-3
P.O. Box 808
Livermore, CA 94550

R. E. Heft
G. H. Higgins
J. B. Knox
P. Phelps
B. W. Shore

2 Los Alamos Scientific
Laboratory
ATT: Report Librarian
P.O. Box 1663
Los Alamos, NM 87544

J. W. Healy
M. F. Milligan

5 Ministry of Agriculture,
Fisheries & Food Laboratory
Lowestoft, Suffolk, England

Librarian

2 National Institute of
Radiological Sciences
Environmental Hygiene Division
4-9-1, Anagawa
Chiba-shi, Japan

M. Saiki
M. Suzuki

5 Oak Ridge National Laboratory
Oak Ridge Operations Office
P.O. Box X
Oak Ridge, TN 37830

S. I. Auerbach
J. A. Auxier
G. D. O'Kelley
E. G. Struxness
J. E. Turner

1 Ontario Hydro
620 University Avenue
Toronto, Ontario, Canada

Library

1 World Health Organization
Case Postale No. 5
CH-1211
Geneva 20, Switzerland

R. L. Dobson

1 World Meteorological
Organization
Geneva, Switzerland

Librarian

22 Individuals - United States

Dr. James R. Arnold
Chemistry Dept.
University of California,
San Diego
La Jolla, CA 92037

Dr. Robert J. Beyers, Director
Savannah River Ecology
Laboratory
University of Georgia
Savannah River Plant
P.O. Box A
Aiken, SC 29801

Distr-4

<u>No. of Copies</u>	<u>No. of Copies</u>
Dr. Wallace Broecker Lamont Geological Observatory Columbia University Palisades, NY 10964	Bernard Manowitz, Head Radiation Division Brookhaven National Laboratory Upton, Long Island, NY 11973
Dr. David D. Dominick Asst. Administrator Office of Categorical Programs Environmental Protection Agency Washington, DC 29460	Dr. Donald G. McCauley Code 304, Michelson Lab. Department of the Navy Naval Weapons Center China Lake, CA 93555
Education and Information Section Health Physics Division Oak Ridge National Laboratory Oak Ridge, TN 37830	Dr. Clarence Menninga Physics Department Calvin College 1331 Franklin St. SE Grand Rapids, MI 49506
Mr. Marvin C. Gaske Asst. to Executive Secretary Advisory Committee on Reactor Safeguards U.S. Nuclear Regulatory Commission Washington, DC 20555	Dr. Dan Montgomery Environmental Protection Agency 26 W. St. Clair St. Cincinnati, OH 45268
Dr. Stanley M. Greenfield Asst. Administrator for Res. and Monitoring Environmental Protection Agency Washington, DC 20460	Dr. Allyn Seymour Lab. of Radiation Ecology Univ. of Washington College of Fisheries Seattle, WA 98105
Mrs. Naomi Harley New York Univ. Medical Center 550 First Avenue New York, NY 10016	Dr. C. W. Sill U.S. Energy Research and Development Administration Idaho Operations Office P.O. Box 2108 Idaho Falls, ID 83401
Mr. H. W. Kirby Monsanto Research Corp. Mound Laboratory Miamisburg, OH 45342	Sam Simmons Environmental Protection Agency Research Triangle Park, NC 27711
Dr. Paul K. Kuroda Dept. of Chemistry University of Arkansas Fayetteville, AR 72701	Mr. Walter Singlevich Air Force Technical Applications Center/TD-4 Patrick AFB, FL 32925
Dr. Philip LaFleur Nuclear Reactor Laboratory National Bureau of Standards Gaithersburg, MD 20760	Dr. Charles V. Theis U.S. Geological Survey P.O. Box 4369 Albuquerque, NM 87106
Mr. Art Levin Battelle Memorial Institute 2030 M Street NW Washington, DC 20545	Dr. Dale Uhl Nuclear Engineering and Operations Dept. Electric Power Research Institute 3412 Hillview Ave. P.O. Box 10412 Palo Alto, CA 94303

Distr-5

No. of
Copies

No. of
Copies

10 Individuals - Foreign

ONSITE

Dr. Thomas M. Beasley
International Laboratory of
Marine Radioactivity
International Atomic Energy
Agency - Oceanographic Museum,
Monaco-Ville,
Principality of Monaco

2 ERDA Richland Operations Office

B. J. Melton
P. W. Gottschalk

Dr. J. C. Dalton
U.K. Atomic Energy Authority
Windscale Works, P. G.
Sellafield, Seascale,
Cumberland
ENGLAND

1 Atlantic Richfield Hanford
Company

D. J. Brown

Dr. K. Edvarson
Forsvarets Forskningsanstalt
Res. Institute of National
Defence
Avdelning 4,
Stockholm, 80, SWEDEN

1 United Nuclear Industries
Richland, Washington

UNI Files

2 Hanford Environmental Health
Foundation

Medical Library

Dr. F. Girardi
C.C.R. EURATOM-ISPRA
(VERESE) - ITALY

117 Battelle-Northwest

T. W. Ambrose
W. J. Bair(5)
N. E. Ballou
J. B. Burnham
N. E. Carter
J. R. Corley
G. M. Dalen
J. M. Davidson
H. Drucker
C. E. Elderkin
S. J. Farmer
W. D. Felix (10)
J. W. Finnigan
R. F. Foster
J. C. Fox (5)
J. J. Fuquay
G. M. Garnant
W. A. Glass (10)
A. J. Haverfield (5)
R. L. Hooper
V. G. Horstman
L. J. Kirby
R. C. Liikala
I. C. Nelson (5)
J. M. Nielsen (20)
D. E. Olesen
J. F. Park
R. W. Perkins (5)
L. A. Rancitelli (15)
W. D. Richmond
C. L. Simpson
W. H. Swift

Dr. Stanley Jackson
U.K. Atomic Energy Authority
Harwell, Didcot, Berkshire
ENGLAND

Prof. D. Lal
Physical Research Laboratory
Navrangpura,
Ahmedabad-9, INDIA

Dr. Alessandro Malvicini
Chief, Protection Service
C.C.R. EURATOM-ISPRA
(VERESE) - ITALY

Dr. J. K. Miettinen
Univ. of Helsinki
Dept. of Radiochemistry
Unionkatu 35, Helsinki
FINLAND

R. V. Osborne
Atomic Energy of Canada, Ltd.
Chalk River, Ontario, CANADA

Dr. D. H. Peirson
AERE (Atomic Energy Res. Estb.)
Health Physics & Med. Div.,
B. 364
Harwell, Didcot, Berkshire
ENGLAND

No. of
Copies

Battelle-Northwest - Continued

W. L. Templeton
R. C. Thompson
C. M. Unruh
B. E. Vaughn
W. R. Wiley
Biology Library (2)
Technical Information (5)
Technical Publications (2) 1 BE