ENVIRONMENTAL MEASUREMENTS LABORATORY
PROGRAM REVIEW — DECEMBER 1983

March 1984

Environmental Measurements Laboratory
New York, New York

Technical Information Center
Office of Scientific and Technical Information
United States Department of Energy

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

This report has been reproduced directly from the best available copy.

Available from the National Technical Information Service, U. S. Department of Commerce, Springfield, Virginia 22161.

Price: Printed Copy A06
Microfiche A01

Codes are used for pricing all publications. The code is determined by the number of pages in the publication. Information pertaining to the pricing codes can be found in the current issues of the following publications, which are generally available in most libraries: *Energy Research Abstracts (ERA)*; *Government Reports Announcements and Index (GRA and I)*; *Scientific and Technical Abstract Reports (STAR)*; and publication NTIS-PR-360 available from NTIS at the above address.

ENVIRONMENTAL MEASUREMENTS LABORATORY
PROGRAM REVIEW - DECEMBER 1983

Editors: Herbert L. Volchok, Gail de Planque

March 1984

ABSTRACT

This volume contains all of the written material that was submitted to the panel of Reviewers in advance of a Program Review conducted by the U. S. Department of Energy, Office of Health and Environmental Research at the Environmental Measurements Laboratory (EML) December 7-9, 1983. In addition to a general introduction there are nineteen papers grouped into the five broad program categories covering all of the scientific and engineering projects of the Laboratory: Natural Radioactivity and Radiation, Anthropogenic Radioactivity and Radiation, Non-nuclear, Quality Assurance, and Development and Support. These short articles, for the most part, focus on the rationale for EML's involvement in each project, emphasizing their relevance to the EML and Department of Energy missions. Project results and their interpretation were presented at the Review and can be found in the material referenced in this volume.

PREFACE

On December 7-9, 1983, a two and one half day review of technical programs was held at the Environmental Measurements Laboratory (EML). The review was requested and administratively organized by EML's parent organization, the Office of Health and Environmental Research (OHER), U. S. Department of Energy. In all there were thirteen reviewers; six were OHER personnel, seven were experts invited by OHER. Following are the reviewers and their affiliations:

Outside Reviewers

Dr. Lynn R. Anspaugh, Lawrence Livermore Laboratory
Dr. Rudolph Englemann, National Oceanographic and Atmospheric Administration
Dr. Jeffrey S. Gaffney, Brookhaven National Laboratory
Dr. Isaac R. Kaplan, University of California at Los Angeles
Dr. William S. Lyon, Oak Ridge National Laboratory
Dr. Vincent Schultz, Washington State University
Dr. Richard Tropp, Texas Air Control Board

OHER Reviewers

Dr. David Ballantine, Division of Physical & Technological Research
Dr. Charles W. Edington, Associate Director, OHER
Dr. Gerald Goldstein, Division of Physical & Technological Research
Dr. Helen McCammon, Director, Division of Ecological Research
Mr. David H. Slade, Deputy Director, Division of Physical & Technological Research
Dr. Robert Watters, Division of Ecological Research

The total Program Review consisted of twenty individual presentations plus laboratory visits for demonstrations and exhibits. The laboratory visits, scheduled for a minimum of one hour each, for groups of eight or less, were for the purpose of allowing and encouraging the reviewers to directly observe a number of EML projects and equipment in an almost "hands on" fashion. The talks and demonstrations were given by the EML personnel engaged in the projects, enabling reviewers to question or discuss design, operation and results in as much detail as they desired in a semi-informal setting. These sessions were also designed for, and indeed resulted in, numerous extended "one-on-one" meetings between EML scientists or engineers and reviewers. The six demonstrations/exhibits were as follows:

Radiation Physics

Mobile Environmental Research Laboratory
Field and Laboratory Radiation Measurements: Equipment, Procedures and Results

Aerosol Studies

Radon and Aerosol Programs
"HATCH" - High Altitude Test Chamber

Environmental Studies

Surface and Upper Air Sampling Operations
Fallout Equipment and Procedures
"Acid Rain"/Freshwater Ecology Operations

Analytical Chemistry

Low Level Radioactivity Laboratories and Counting Equipment
"Acid Rain" Analytical Facilities
Clean Room

Instrumentation

Equipment Design, Engineering and Fabrication Facilities
On-line Data Acquisition Systems

Applied Mathematics

VAX-11/750 Computer - Graphics, Information Retrieval, and Interactive
Data Management

The twenty presentations, except for the introduction, were organized into five major categories covering virtually all EML technical activities.

- Anthropogenic Radioactivity and Radiation Programs
- Natural Radioactivity and Radiation Programs
- Non-nuclear Programs
- Quality Assurance Programs
- Development and Support Programs

To facilitate the review process EML distributed to the Reviewers a paper corresponding to each presentation prior to the actual meeting. Each paper focused first on the rationale for doing the particular project and provided an explanation of its history and background. Secondly each emphasized the relevance of the project to the Department and the Laboratory and finally discussed the future directions of the work. To keep the papers concise, results, data tables and figures were omitted; references and attached reprints provided these details.

This volume contains the twenty papers, distributed to the Reviewers.

TABLE OF CONTENTS

	<u>Page</u>
<u>Introduction</u> - H. L. Volchok and G. de Planque	1
 <u>Anthropogenic Radioactivity and Radiation Programs</u>	
Global Fallout - E. P. Hardy, R. Larsen, C. Klusek	5
Special Dosimetry - K. O'Brien	16
Radiological Impact of Nuclear Facilities - C. Gogolak	24
Reconstruction of Fallout Exposure from Nevada Weapons Testing - H. Beck	24
 <u>Natural Radioactivity and Radiation Programs</u>	
Radiological Impact of Alternate Energy Sources - C. Gogolak . . .	33
Environmental Radiation - K. Miller	37
Natural Radioactivity in the Atmosphere	42
Radon Programs - A. George	45
 <u>Non-Nuclear Programs</u>	
Atmospheric Tracer Studies - R. Lagomarsino	51
Aerosol Programs - E. O. Knutson	54
Pollutants in the Upper Atmosphere - R. Leifer	60
Atmospheric Deposition - H. Feely, D. Bogen	65
Energy-Related Pollutants in Ecosystems - M. Heit	71
 <u>Quality Assurance Programs</u>	
Quality Assurance Programs at EML-Overview - H.L. Volchok . .	82
The EML-OOS Quality Assessment Program - C. Sanderson	85
Radon Intercomparisons - I. M. Fisenne	87
TLD Intercomparisons - G. de Planque	91
 <u>Development and Support Programs</u>	
Instrumentation Development and Support Programs - R. T. Graveson	96
Analytical Support and Development - P. W. Krey	104

INTRODUCTION

Herbert L. Volchok
Gail de Planque

The Environmental Measurements Laboratory (EML) is engaged in energy related research for the purpose of addressing general and specific problems of interest to the DOE. EML is responsible for evaluating the exposure of humans to environmental radiation, radioactivity and other toxic materials associated mainly with energy production. The Laboratory develops and improves techniques for studying pathways of contaminants from source to people and applies new technology developments to the detection, analysis and assessment of ionizing radiation, trace substances and other pollutants.

The Laboratory can be first identified as a clearly distinct unit in 1947, as the Medical Division of the New York Operations Office (NYO) of the Atomic Energy Commission. Technical direction of the programs was transferred to AEC Headquarters under the Division of Biology and Medicine (now known as OHER) in 1952 and the name was changed to the Health and Safety Laboratory one year later, a name we kept for twenty three years. In 1972 NYO was closed and the administrative responsibility for EML went to AEC Headquarters only to be transferred again to Chicago Operations Office in 1981. Always remaining a part of OHER, we went through two years under the Energy Research and Development Administration (ERDA) and then as part of the Department of Energy (DOE) in 1981 we were put under the Office of Energy Research.

This thirty six year record of the Laboratory testifies to its proper role as a government facility, providing three features which can neither be delegated to nor expected from non-public organizations: (a) responsiveness (b) continuity and (c) status.

- (a) EML responds immediately and authoritatively to requests from Headquarters without negotiation, qualification or delay. Through the years EML has responded on numerous occasions involving environmental pollution incidents which could have been hazardous, embarrassing and costly.
- (b) Since the early 1950's, EML has studied world-wide radioactivity and radiation as well as non-radioactive substances, directed toward understanding

the environmental pathways to and impacts on humans. These include the only continuous and systematic observations of nuclear debris in the upper atmosphere and at the earth's surface in the world. The Laboratory has made outstanding contributions through these programs, including studies of nuclear fallout, of U. S. and Russian satellite failures and of stratospheric ozone destruction by freons and other chlorocarbons. Such networks have been maintained for several decades under the aegis of the federal government and EML.

- (c) As a U. S. Government facility involved in research of national or international scope, EML's status provides access to representatives of foreign governments and international organizations on a "government-to-government" basis. This authority greatly facilitates the types of cooperative, inter-agency, inter-governmental programs which are absolutely necessary for successful, large scale environmental research in the national interest.

In carrying out its mission, EML plans, directs and executes:

- 1) studies of the distribution, composition, mechanism and rate of movement of nuclear and non-nuclear pollutants from the atmosphere to soils, plants, animals and humans,
- 2) radiological and chemical monitoring programs to develop data used by DOE, other Federal agencies, and international organizations,
- 3) advanced studies in the development and application of analytical chemistry and radioanalytical procedures,
- 4) environmental research studies,
- 5) research and development in the area of instruments for use in detecting and measuring environmental radioactivity and non-nuclear pollutants, and
- 6) a variety of quality assurance activities ranging from "in-house" to intra and inter agency cooperative programs to international intercomparisons.

In addition the Laboratory:

- 7) provides environmental health radiation physics, and environmental contamination control surveys for specific problems of interest to the department, and
- 8) provides technical assistance to Headquarters' Divisions as requested through the Office of Health and Environmental Research.

EML occupies the entire fifth floor, some 75,000 square feet, of a 12 story government-owned building in the southwest part of Greenwich Village, in New York

City. The Laboratory has been at this location for 24 of its 36 year history as a government facility under A. E. C. and E. R. D. A. before DOE.

Of the 93 (full time equivalent) employees at this time, 76% are technical and 57% are professional. The professional staff consists of specialists in: organic/inorganic/geo/radio-chemistry, radiation/aerosol physics, biology, ecology, meteorology, mathematics, computer science and mechanical/electrical engineering. State-of-the-art laboratories and equipment in many of the above named disciplines have been developed or obtained over the years and a number of very specialized facilities have been made available.

In rural Chester, N. J., about 40 miles due west of the Laboratory, EML operates a field station for the purpose of carrying out experiments and observations on regional pollutant levels as well as providing a more general environmental setting than New York City for testing and calibrating field instruments. Chester is also being utilized as one of the important, instrumented locations for the Cross Appalachian Tracer Experiment (CAPTEX).

The EML work boat is a unique 22' x 12' catamaran which can be transported disassembled by auto-towed trailer. Since it was obtained in 1978, more than 20 lakes in the United States have been studied, sampling water, biota and about 100 large diameter (24 cm.) sediment cores of up to one meter in length. Careful analysis for ^{137}Cs , ^{210}Pb and plutonium allows the assigning of dates of deposition to individual sediment layers, an invaluable factor in studying the history of various atmospheric pollutants.

The EML mobile radiation laboratory is almost unmatched in being able to provide accurate and precise in-situ, real-time germanium diode gamma spectrometry in the field as well as other associated measurements typically required in environmental dose studies such as Nevada-Utah or Three Mile Island.

The radon chambers at the Laboratory can be accurately controlled over wide concentration ranges of radon for calibrating, testing and intercomparing instruments.

The High Altitude Test Chamber (HATCH) is a large specially constructed room, capable of reproducing temperatures and pressures in excess of altitudes of 90 kilometers,

used for calibrating equipment involved in the Laboratory's environmental programs, from sea level into the stratosphere.

One additional facility at the Laboratory which is undoubtedly unique is the extensive library of samples, going back in some cases 30 years or more. The sample types include human bone, animal bone and tissue, vegetation, milk and food ash, soil, lake sediment and water, seawater and filter paper ranging in altitude from sea level to the stratosphere. In all, this archive consists of more than 45,000 samples.

The Laboratory owns a VAX-11/750 (DEC) Computer with the necessary peripheral equipment to provide essentially state-of-the-art capability in data processing and associated functions.

The principal "product" of the Laboratory consist of the reports of the results of the research programs. This has been a gradually increasing function over the past 20 years. In recent years the mean rate of publications has been over 60 articles per year about 20 of which appear in recognized (peer reviewed) scientific journals. In addition the staff averages in excess of 40 scientific presentations per year.

THE GLOBAL FALLOUT PROGRAM

Edward P. Hardy
Richard Larsen
Catherin S. Klusek

Introduction

The global fallout program, as it currently operates at the Environmental Measurements Laboratory (EML), consists of a number of related investigations involving periodic or continuous sampling and measurements of radionuclides. The sources of these radionuclides are primarily nuclear weapons tests which inject radioactive debris into the atmosphere. DOE, like the AEC before it, has a responsibility for gathering information on this debris. Included in the program are measurements of air filters collected by aircraft and balloons in the upper atmosphere, of air filters collected in surface air, of wet plus dry deposition, and finally, of human foods and bone tissue. Our expertise in each of these areas of investigation is unique because of the number of years that measurements have been made, the uninterrupted nature of the sampling and the information provided.

History and Objectives

The Laboratory first became involved in the measurement of long-range radioactive fallout in 1951 following evidence that fission products from explosions at the Nevada Test Site were being distributed across the country (Harley, 1976). Studies began at EML, then the Health and Safety Laboratory, and at other laboratories after concern had been expressed over possible health hazards from long-range fallout. At the time the most critical radionuclide was considered to be ^{90}Sr . In retrospect it would have been useful to include measurements of other radionuclides in addition to ^{90}Sr , but uncertain information and inadequate instrumentation precluded investigations of ^{137}Cs , ^{131}I and Pu isotopes, for example. Over the next several years the Laboratory developed a nationwide, and eventually a worldwide fallout deposition network involving simple collection systems.

Initially the network used gummed-film to collect fallout particles, which were counted for total beta activity (Harley et al., 1960). From this, calculations were made of ^{90}Sr deposition and the external gamma dose. This system was discontinued in 1959 as methods were developed to analyze the total deposition in a stainless steel pot collector directly for ^{90}Sr . By 1958, 32 stations in the U. S. were collecting monthly fallout for the Laboratory, and by 1960 we had established over 100 stations worldwide. This was largely made possible through the cooperation of the U. S. State Department, the U. S. Weather Bureau and scientists in other countries, and the successful development and deployment of a collection system which uses a plastic funnel and an ion exchange column. This method allowed people untrained in scientific techniques to collect and return samples to us with little effort.

Over the years it has often been demonstrated that ^{90}Sr can be used as a tracer for long-lived fission products in general and that it provides a background for placing nuclear contamination from other sources into perspective (Hardy et al., 1962; Volchok, 1964; Volchok, 1966; Volchok, 1967; Volchok and Krey, 1967). The rationale for ^{90}Sr measurements still holds, although the relative importance of this radionuclide as it pertains to human exposure has diminished somewhat. Since it is an alkaline earth element which imitates Ca in biological systems, the eventual retention of ^{90}Sr in bone tissue makes it a critical radionuclide in delivering a dose to the human. Prompted by public concern over the possible health effects of worldwide fallout from large scale nuclear tests, the Laboratory has, since 1960, also periodically measured the amounts of ^{90}Sr in human diet and bone (Klusek, 1982a; Klusek, 1982b). Because of its long physical and biological half-life, ^{90}Sr continues to be the most persistent artificial radionuclide which reaches man through ingestion. Soil is not the ultimate sink for ^{90}Sr as it is for other long-lived fission products. Rather, ^{90}Sr has a propensity for moving into plants via root systems, and consequently remains in easily measurable amounts in human food and bone (Rivera, 1963; Rivera, 1965; Hardy and Rivera, 1968; Rivera, 1968; Hardy and Bennett, 1977). Presently, ^{90}Sr contributes about 16 percent of the internal dose to the total body and about 30 percent of the total dose to bone surfaces and marrow.

It was during the 1957 Congressional Hearings on Fallout that the responsibility of the Laboratory to track nuclear debris on a global scale became abundantly clear. Until that time much of the information on ^{90}Sr exposure was classified and conjectural, and public distrust was beginning to mount. From that point on, the policy of the Laboratory has been to rapidly publish all data on radioactive fallout. The Fallout Program Quarterly Summary Reports, and recently the Environmental Reports published by the Laboratory have been dedicated to releasing information as quickly as possible and to providing background and interpretive material to the scientific community (Larsen, 1983).

These early investigations of the Laboratory continue today, although at a somewhat reduced level of effort. The ^{90}Sr global fallout network and the human diet and bone studies still provide basic information required to assess human exposure from nuclear tests. The fallout network is the only one in existence from which an inventory of the global deposit at any point in time can be derived (Larsen, 1983; HASL-329, 1977; Volchok and Kleinman, 1969; Feely et al., 1978). This network, backed up by the continuous measurements of ^{90}Sr in diet and bone tissue, provides the framework which has enabled the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) to model the relationships among deposition, diet and bone and to provide reliable estimates of human exposure (UNSCEAR 1982).

Air sampling at ground level and in the stratosphere, which began about the same time as the ^{90}Sr fallout, diet and bone investigations, is also a coherent part of the Laboratory's global fallout program. Initially there was considerable uncertainty regarding the critical pathway for human exposure, and the Naval Research Laboratory established an air filter sampling network extending from Northern Canada to the tip of South America. In 1963, the Laboratory took over this network and expanded it to include both the Arctic and Antarctic regions. Although it eventually became clear that human exposure via inhalation was relatively insignificant for long range ^{90}Sr fallout, surface air sampling has been continued and has proven its scientific as well as political worth (Volchok, 1965; Kleinman and Volchok, 1969; Feely et al., 1981). The amount of air sampled is large enough to measure other radionuclides such as plutonium whose major exposure pathway to people is via inhalation (Bennett, 1974; Bennett, 1975). It is the only

operating system that can signal the arrival of fresh debris from nuclear tests and can indicate the hemispheric distribution of that debris with time. The air filters can be counted within a reasonably short time after sampling. This has made this network on numerous occasions, the primary source of verification information.

Finally, an integral tie-in to the Laboratory's capability to track nuclear test debris is the High Altitude Sampling Program (HASP)(HASP Brochure, 1980). When it became obvious that thermonuclear tests, starting with IVY-MIKE in 1952, carried radioactive particles well up into the stratosphere, no one knew for sure how long it would take for this debris to eventually fall out. Originally it was hypothesized that the stratospheric residence time was on the order of ten years, which would minimize the eventual exposure of people to fission products. The first stratospheric measurements were made under the Air Force ASHCAN balloon program in 1956 for which the Laboratory took over the analytical phase in 1961. Subsequently aircraft sampling was initiated by the Defense Atomic Support Agency. In 1967 the Laboratory was assigned responsibility for both sampling programs and named the aircraft program AIRSTREAM. As data became available it was found that the average residence time was only about a year; slightly longer for equatorial tests and shorter for arctic tests (Krey and Krajewski, 1970). This information drastically increased the perceived importance of the contribution of large-yield explosions to human exposure. The use of balloons and aircraft in conjunction with the deposition network enabled the Laboratory to maintain a balanced inventory of ⁹⁰Sr. Knowledge of the temporal distribution and isotopic composition of stratospheric nuclear debris allowed the development of models of stratospheric transport and transfer of material to the surface of the earth (Krey, 1967). It became possible to predict deposition patterns in advance, which made estimates of human exposure more realistic. Subsequent analyses of ⁹⁰Sr in soil verified the predicted integrated deposition patterns (Hardy et al., 1968; Hardy et al., 1972).

With this historical background in mind it is now appropriate to describe briefly the current global fallout program in operation at the Laboratory.

The ^{90}Sr Deposition Network

Today the 65 stations collecting total deposition throughout the world represent the minimum geographic coverage needed to determine annual ^{90}Sr fallout fluxes (Larsen, 1983; Hardy, 1975; Toonkel, 1981). Many of these stations have provided samples to the Laboratory continuously for 25 years. In recent years nuclear tests by the Peoples Republic of China have contributed the major proportion of the measurable background rates of ^{90}Sr fallout (Telegadas, 1979). When unexpected weather patterns have caused unusually heavy rain-out of nuclear debris, as occurred over the eastern U. S. in 1976, this global network was invaluable in assessing the impact of this event on population exposures. Were this network discontinued, it would be virtually impossible to resurrect such a network in the event of unforeseen large-scale nuclear contamination.

^{90}Sr in Human Diet and Bone

Over the years covered by the nuclear fallout era there have been many measurements made of ^{90}Sr in food and human tissue. The investigations at the Laboratory have led to the development of models that can reasonably correlate deposition with concentrations in food and finally body burdens of ^{90}Sr (Bennett, 1972; Klusek, 1982a; Klusek, 1982b). Quarterly samplings of food categories in New York and San Francisco have provided a long-term record of ^{90}Sr intake for these typical Northern Hemisphere population regions. The fact that ^{90}Sr fallout has consistently been higher in New York than in San Francisco has been reflected in the record of food and bone tissue concentrations. This has allowed the dependence of bone concentration on dietary intake to be understood in a quantitative way, and has led to mathematical expressions to describe the extent of absorption and retention in the human (Rivera, 1965). Clinical experiments with human subjects carried out on a cooperative basis with the Hines Veterans Hospital in Chicago have provided verification of these derived relationships (Spencer et al., 1963; Spencer et al., 1964; Spencer et al., 1967; Spencer et al., 1968; Spencer et al., 1972; Spencer et al., 1973; Kramer et al., 1973; Spencer et al., 1977).

Since ^{90}Sr remains at easily measurable levels in diet and bone, it is the intention of the Laboratory to continue this investigation of the long-term behavior of ^{90}Sr in the

environment and of its transfer to people. Other scientific groups continue to use the Laboratory data and models of deposition to diet and to bone confirming the current relevance of the program (Toggweiler, 1983; Legett, et al., 1982; Pepworth and Vennart, 1973; Ing, 1983; ICRP 1972; WASH-1405, 1975).

Surface Air Sampling

Through the cooperation of scientists in this country, Canada, and countries in South America, the Laboratory is able to collect filter samples of surface air at 22 stations covering the latitude range from 71°N to 90°S. The volume of air sampled is large enough to allow the direct measurement of the filters by gamma spectrometry for a number of radionuclides of varying half-life, such as ^7Be , ^{95}Zr , ^{137}Cs , ^{144}Ce , ^{140}Ba and ^{131}I , and by radiochemistry for ^{90}Sr and $^{239,240}\text{Pu}$. This capability has placed the Laboratory, on a number of occasions, in the position of providing the only definitive information relating to an uncontrolled or unexpected nuclear event. One example of this was the Vela satellite observation of a possible nuclear explosion in the South Atlantic in September 1979. The Laboratory found no evidence of fresh debris in any of the filtered air samples analyzed at that time, which, based upon many years of observations of atmospheric nuclear events in both hemispheres, suggested that no such explosion had occurred.

High Altitude Sampling

Balloon and aircraft sampling of particulate radioactivity has provided one of the major data bases for predicting fallout patterns and calculating dose commitments to people from nuclear tests, using models developed by UNSCEAR.

Normally, balloons are launched three times a year at Alamogordo, NM to coincide with aircraft sampling. NASA flies the airplane for DOE during the spring, summer and fall seasons in the Northern Hemisphere along tracks extending from 75°N to the equator. Four altitudes are sampled ranging from 12 km to 19 km. As in the surface air program, these air filter samples are analyzed by gamma spectrometry for short and long-lived gamma emitters, and by radiochemistry for ^{90}Sr and $^{239,240}\text{Pu}$. The mass isotopic composition of plutonium in selected samples is measured to characterize the nuclear debris in the stratosphere.

There have been several specific instances where it has been possible to evaluate the impact of unplanned nuclear releases because of this high altitude sampling capability. For example, following the destruction of a SNAP-9A satellite in 1964, it was possible to determine the amount of ^{238}Pu released from the power source and to show that complete burn-up had occurred (Krey, 1967). This information was used to plan a ground sampling investigation which allowed the Laboratory to provide the distribution pattern and inventory of global contamination from this nuclear event (Hardy et al., 1973; Harley, 1980; Volchok et al., 1983). More recently, when the Soviet satellite Cosmos-954 re-entered the atmosphere over Northern Canada, the Laboratory was able to sample the atmosphere to an altitude of 39 km, and to detect particulate debris from the satellite reactor (Krey et al., 1979). The finding that the Cosmos reactor had partially burned-up in the upper stratosphere explained why the ground surveys could not account for all of the radioactive components. A similar investigation of the second Soviet satellite, Cosmos 1402, which re-entered the atmosphere in January of this year is possible but the Laboratory is on standby pending funding and deployment opportunities.

The Future

Implied in the foregoing discussion is the Laboratory's position that the Global Fall-out Program should continue, as presently operated, indefinitely. Whenever the public becomes apprehensive about nuclear testing it seems that the Laboratory sampling networks are expected to be in operation and responsive.

Sir William Pochin of the British National Radiological Protection Board was asked by Lauriston Taylor in a recorded interview why the United Nations Scientific Committee on the Effects of Atomic Radiation should remain in existence in view of diminished fallout levels and concomitant diminished anxiety associated with fallout. His reply was that it was far better for UNSCEAR to remain a viable scientific body in case of a recurrence of fallout and/or a recurrence of anxiety, for it would be impossible to muster such a prestigious body of scientists in time to provide the public with the exposure information needed to make rational judgments. Likewise the Laboratory feels that the on-going investigations of global fallout represent a national resource for providing information in an uncertain nuclear future.

References

- Bennett, B., Estimation of ^{90}Sr Levels in the Diet, USAEC Report HASL-246, p I-107 (1972)
- Bennett, B., Fallout ^{239}Pu Dose to Man, USAEC Report HASL-278, p I-41 (1974)
- Bennett, B., Transuranic Element Pathways to Man, IAEA, SM-199/40 (1975)
- Feely, H., H. Volchok, E. Hardy and L. Toonkel, Worldwide Deposition of ^{90}Sr Through 1976, Environ. Sci. & Tech. 12, 808-809 (1978)
- Feely, H., L. Toonkel and R. Larsen, Radionuclides and Trace Elements in Surface Air, USDOE Report EML-395, pp C-1-C-205 (1981)
- Hardy, E. P. and L. Alexander, Rainfall and Deposition of Strontium-90 in Clallam County, Washington, Science 136, 881-882 (1982)
- Hardy, E. P. and J. Rivera, Transfer of Fallout ^{90}Sr to Cows' Milk, J. Dairy Sci, 51, 1210-1214 (1968)
- Hardy, E. P., M. Meyer, J. Allen and L. Alexander, Strontium-90 on the Earth's Surface, Nature 219, 584-587 (1968)
- Hardy, E. P., H. Volchok and P. W. Krey, Strontium-90 Fallout in Utah, USAEC Report HASL-257 (1972)
- Hardy, E. P., P. W. Krey and H. Volchok, Global Inventory and Distribution of Fallout Plutonium, Nature 241, 444-445 (1973)
- Hardy, E. P. and B. Bennett, Radionuclide Uptake by Cultivated Crops, USERDA Report HASL-321 (1977)
- Hardy, E. P., Regional Uniformity of Cumulative Radionuclide Fallout, USAEC Report HASL-288 (1975)
- Harley, J. H., N. Hallden and L. Ong, Summary of Gummed Film Results Through December 1959, USAEC Report HASL-93 (1960)
- Harley, J. H., A Brief History of Long-Range Fallout, USERDA Report HASL-306, pp I-3 - I-19 (1976)
- Harley, J. H., Plutonium in the Environment - A Review, J. Radiat. Res. 21, 83-104 (1980)

- HASL-329, Final Tabulation of Monthly ^{90}Sr Fallout Data: 1954-1976, USERDA Report HASL-329 (1977)
- HASP Brochure, Summary of the High Altitude Sampling Program, USDOE, Environmental Measurements Laboratory, New York (1980)
- ICRP, Bone and Food Data used by Task Group on Local Retention Functions of Bone Seekers in Study of Retention Function of Compact bone, Cancellous Bone, Bone Surface, Soft Tissue, Blood Plasma and Whole Body (1972)
- Ing, Y, Dose Assessment Model for Ingestion of 20 Nuclides: Calculation of Dose Factor for ^{90}Sr in Adult Bone used EML Bone Model (1983)
- Kleinman, M. and H. Volchok, Direct Relationship of Surface Air Radionuclide Concentrations to Global Fallout, Science 166, (1969)
- Klusek, C., Strontium-90 in the Diet - Results through 1981, USDOE Report EML-412 (1982a)
- Klusek, C., Strontium-90 in Human Bone: 1981 Results for New York City and San Francisco, USDOE Report EML-412 (1982b)
- Kramer, L., N. Spencer and E. Hardy, Dietary Strontium-90 Intake in Chicago, Health Phys. 25, 445-448 (1973)
- Krey, P., Strontium-90 Concentrations and Stratospheric Transport, J. Geophys. Res., 72, 2987-2996 (1967)
- Krey, P., Atmospheric Burn-up of a Plutonium-238 Generator, Science 158, 769-771 (1967)
- Krey, P. and B. Krajewski, Comparison of Atmospheric Transport Model Calculations With Observations of Radioactive Debris, J. Geophys. Res., 75, 2901-2908 (1970)
- Krey, P., R. Leifer, W. Benson, L. Dietz, H. Hendrickson and J. Coluzza, Atmospheric Burn-up of the Cosmos-954 Reactor, Science, 205, 583-585 (1979)
- Larsen, R., Worldwide Deposition of ^{90}Sr Through 1981, USDOE Report EML-415 (1983)
- Legett, R., K. Eckerman and L. Williams, Strontium-90 in Bone: A Case Study in Age-Dependent Dosimetric Modeling, Health Phys. 43, 307 (1982)
- Papworth, D. and J. Vennart, Retention of ^{90}Sr in Human Bone at Different Ages and the Resulting Radiation Doses, Phys. Med. Biol. 18, 169 (1973)

- Rivera, J., Strontium-90 in Human Diet and Bone, *Radiology*, 80, 120-123 (1963)
- Rivera, J., Human Bone Metabolism Inferred from Fallout Investigations, *Nature* 207, 1330-1332 (1965)
- Rivera, J., Factors Affecting the Uptake and Retention of Bone-Seeking Radionuclides, *Pædiatrics*, 41, 231 (1968)
- Spencer, H., J. Samachson, E. Hardy and J. Rivera, Strontium-90 Metabolism in Man, *Clinical Science* 24, 405 (1963)
- Spencer, H., J. Samachson, E. Hardy and J. Rivera, Strontium-90 Metabolism During Low Strontium-90 Intake in Man, *Radiation Res.* 22, 668-676 (1964)
- Spencer, H., J. Samachson, E. Hardy and J. Rivera, Effect of Low and High Calcium Intake on Strontium-90 Metabolism in Adult Man, *Intl. J. Appl. Rad. & Isotopes* 18, p. 605 (1967)
- Spencer, H., J. Samachson, E. Hardy and J. Rivera, Some Aspects of the Effects of Calcium Intake and Age on Strontium-90 Metabolism in Man, *Health Physics* 15, 499-504 (1968)
- Spencer, H., J. Samachson, E. Hardy and J. Rivera, Effect of Orally and Intravenously Administered Stable Strontium on ^{90}Sr Metabolism in Man, *Rad. Res.* 51, p. 190 (1972)
- Spencer, H., L. Kramer, J. Samachson, E. Hardy and J. Rivera, Strontium-90-Calcium Interrelationships in Man, *Health Phys.* 24, 525-534 (1973)
- Spencer, H., L. Kramer and E. Hardy, Effect of Phosphorous on the ^{90}Sr Balance in Man, *Health Phys.* 22, 417-423 (1977)
- Telegadas, K., Radioactivity Distribution in the Stratosphere from the Chinese High Yield Nuclear Test of November 17, 1976, USDOE Report EML-356 (1979)
- Toggweiler, J. R., A Multi-Tracer Study of the Abyssal Water Column of the Deep Bering Sea Including Sediment Interactions -- A Six Zone Refinalized Model for Bomb Radiotracers and CO_2 in the Upper Kilometer of the Pacific Ocean, PhD Thesis, Columbia University, New York (1983)
- Toonkel, L., Regional Representativeness of ^{90}Sr Deposition Collections, *Health Phys.* 41, 823-825 (1981)
- UNSCEAR, Ionizing Radiation: Sources and Biological Effects, United Nations Scientific Committee on the Effects of Atomic Radiation (1982)

- Volchok, H. L., Strontium-90: Estimation of Worldwide Deposition, Science 145, 1451-1452 (1964)
- Volchok, H. L., Distribution of Strontium-90 in Surface Air During 1963, Nature 206, 1031-1033 (1965)
- Volchok, H. L., The Global Strontium-90 Budget, J. Geophys. Res. 71, 1515-1518 (1966)
- Volchok, H. L., Strontium-90 Deposition in New York City, Science 156, 1487-1489 (1967)
- Volchok, H. L. and P. W. Krey, Implications of Changes in the Global Strontium-90 Budget, J. Geophys. Res. 72, 5155-5156 (1967)
- Volchok, H. L. and M. Kleinman, Strontium-90 Yield of the 1967 Chinese Thermo-nuclear Explosion, J. Geophys. Res. 74, 1694-1696 (1969)
- Volchok, H. L., P. Krey and J. Holland, Nuclear Power in Space, Science 220 (1983)
- WASH-1405, Food Data and Models Used in Study of Transfer Rates from Soil to Root Systems and Incorporation into Plants and Studies of Milk Contamination (1975)

SPECIAL DOSIMETRY

Keran O'Brien

The five dosimetry programs described below are examples of the extent of the Laboratory's capabilities in the field of radiation dosimetry. These studies range in physical scale from millimeters to thousands of kilometers, and involve the physics appropriate to many different particle types and particle energies. All but the last of these studies are new, but all of them are based solidly on EML's years of experience and engagement in problems associated with the physics of the radiation environment, reactors and particle accelerators.

1) Dosimetry of Choroidal Melanoma Radiotherapy

Because of our considerable experience in radiation transport and because we had earlier carried out an extensive campaign of human organ dose calculations (O'Brien and Sanna, 1976; O'Brien and Sanna, 1978; O'Brien, 1980) we are collaborating with the Department of Radiation Oncology of the Hackensack Medical Center in New Jersey in a study on the dosimetry of the radiation sources used in radiotherapy of the eye.

Malignant melanoma of the choroid is the most common eye cancer. The classical treatment for this cancer is the removal of the eye. Radiotherapeutic treatment has achieved comparable survival rates while saving the eye and at least some of its sight. Because of a higher RBE and because the softer radiations facilitate the protection of hospital personnel, ^{125}I has been substituted for ^{60}Co in these treatments.

The commercially manufactured ^{125}I sources, widely used in radiotherapy are quite small: two 0.06 cm diameter resin spheres inside an 0.45 cm long titanium capsule. As the sources are applied directly to the area of the melanoma by a surgical procedure, the dose rates of interest occupy a small region very close to the source (the eye is only 2.4 cm in diameter), making experimental dosimetry very difficult. Since for radiotherapy it is important to know the delivered dose rates accurately, recourse to radiation transport calculations was necessary.

We have calculated dose distributions from both ^{60}Co and ^{125}I in a three-dimensional approximation to the human head from idealized point sources, (Chiu-Tsao et al., 1982). We are now calculating the radiation distributions around the titanium capsules in detail in a large water medium. In the future we will take into account the irradiation pattern from a distribution of capsules as are actually utilized in the irradiation procedure, and represent the human head more exactly.

The data developed from these calculations will be of direct use to the radiotherapy community and will give us more experience in the use of combinatorial geometry techniques to represent three-dimensional objects for computational purposes.

2) Solar Neutrinos

The sun is central to any consideration of global or regional climatic effects, of meteorology, and of the production of cosmogenic isotopes such as ^{10}Be , ^7Be , tritium or ^{14}C . However, measurements of the solar neutrino flux (Davis et al., 1983) have demonstrated just how little we know about the processes that drive the sun. Careful and detailed calculations of the expected neutrino flux, based on the best hydrodynamical and nuclear theory yield four times the actual measured data.

Recently, Davis discovered that his measurements of the neutrino flux were higher during large solar flares and appeared to be anti-correlated with solar activity as measured by the sunspot number. This raised the possibility that cosmogenic neutrino emitters in the earth's atmosphere produced by cosmic-ray or flare particle bombardment make a significant contribution to the measurements, which would mean that the measured solar neutrino flux was even smaller than had been thought.

This question was explored using theoretical methods because we had developed a reputation over many years of effort expended on developing the theory of radiation transport as applied to atmospheric cosmic rays. We have a unique capability to compute cosmic-ray particle fluxes and interactions in the atmosphere taking into account, among other things, solar activity, geomagnetic phenomena, latitude and altitude.

The results of these calculations showed that neutrinos of tellurian origin are way too few to account for the observed phenomena. The observations remain, however and

we are commencing to explore the possibility that neutrinos from atmospheric cosmic-ray pion, kaon and muon decay can account for these phenomena.

The standard model of the sun's activity can be brought into closer agreement with experiment if various other physical processes are introduced into the theoretical picture of the sun's behavior. Processes that would lower the calculated value of the internal temperature, for example would depress the calculated neutrino flux. Another possibility to be considered is the possibility of neutrino oscillations. A further reduction of the measured values of the neutrino flux might rule out some of these possibilities and point to others.

These calculations of the contribution of cosmic-ray neutrinos may provide data which will help to understand the behavior of the sun.

3) Mixed Field Beta-Gamma Dosimetry

The dosimetry of mixed beta-gamma radiation fields at the applied health physics level is a currently significant but difficult and largely unsolved problem within the nuclear community. Furthermore, the problem has often gone unrecognized at the operating level and many applied health physicists have assumed over the years that field surveys and personnel dosimeter results represented the actual conditions to an acceptable degree of accuracy. This erroneous assumption often led to over-exposures of personnel and resulted in lost working time.

There are numerous places in the nuclear industry where intense high-energy mixed beta-gamma radiation fields can be encountered:

- a) during the maintenance of nuclear fuel reprocessing plants,
- b) during the routine maintenance of nuclear reactors,
- c) during the repair of the steam generators of pressurized water reactors, and
- d) during nuclear reactor refueling operations,

to mention just a few. In addition to the nuclear industry the extensive use of radioactive sources in other industries, medicine and research often require the radiation dose assessment of mixed radiation fields for radiation protection purposes.

EML's experience in beta-ray dosimetry dates from its early history as a division of the New York Operations Office which at that time was part of AEC's Production Division.

The Operations Office had to deal with uranium both in the form of metal and in the form of oxides and fluorides and one of the decay products of uranium, UX₂, is a potent beta emitter. Both theoretical and mensurational programs arose from that experience and led to our being called upon by the Idaho Operations Office for help with a problem that arose at INEL's Waste Calcining Facility in the mid 1970's. To provide them and the nuclear industry with a reliable survey meter, we designed, developed and built a new dosimeter for them which is now used throughout the nuclear industry.

More recently, OHER and EML sponsored an informal beta-ray dosimetry workshop, at which the attendees discussed current beta radiation problems and made specific recommendations including improved health physicist education, provide calibration sources and techniques and develop new measurement techniques and instruments.

To implement these recommendations of the beta-ray dosimetry workshop, in collaboration with LANL and LLNL, we started:

- a) transport calculations to characterize radiation fields,
- b) development of a field spectrometer/dosimeter, and
- c) development of on line unfolding techniques to obtain in-situ spectra.

The primary purpose of the collaboration is the development of a spectrometer-survey instrument. It is expected however, that these developments will help in the design of personnel dosimeters, their calibrations, and the interpretation of the results of such measurements. The calculations will also help to improve applied field procedures and will narrow the large discrepancies between estimated (from field survey measurements) and actual personnel dosimetry data.

EML's part in the collaboration is to provide the basic physics of the radiation detector design and response characteristics, perform the radiation transport calculations, develop measurement, calibration and unfolding techniques, and interpret the results.

This program will provide a very significant contribution by EML to an increasingly recognized problem area and through this program EML can also provide dose equivalent conversion factors and other data to international bodies such as ICRP.

4) A New Type of Moderate-and-Capture Neutron Spectrometer

At many research and energy producing facilities neutrons contribute significantly to personnel radiation exposure. During a recent EML study of the radiation field inside

reactor containment vessels, for instance, the ratio of the neutron to gamma dose-equivalent rates was found to range from 0.1 to as high as ten, and the dose-equivalent rates themselves on occasion were of the order of 50 rem per hour. Thus it is clearly important to monitor this neutron field for radiation protection purposes. Such monitoring is complicated by the energy dependence of the neutron monitors, which is, generally, very poor by comparison with, say, gamma-ray monitors, so that a knowledge of the differential energy spectrum of the neutrons being monitored is necessary to interpret their response.

The Environmental Measurements Laboratory has successfully used the multisphere spectrometer system to measure neutron spectra for many years (O'Brien et al., 1965). Our involvement grew out of the fact that the Department of Energy and its predecessor agencies owned many of the research facilities and helped sponsor the development of nuclear energy and yet had no "in-house" capabilities. We were urged to develop these capabilities in the early 50's which led to an extensive program of dose and flux measurements, shielding calculations and neutron spectrometry.

The most recent application of these capabilities was to the measurement of neutron spectra in reactor containment vessels mentioned above. It was a time of some controversy, when it was alleged in the media that the atomic energy program was in considerable difficulty as the neutron fluxes to which reactor operating personnel were exposed had an energy distribution that could not be measured by current instrumentation. Twenty-six measurements were made in seven different PWR containment vessels (Sanna et al., 1982).

The moderate-and-capture type of spectrometer is the only one with the sensitivity and portability necessary for radiation protection purposes that covers so wide an energy range (from thermal to a few hundred MeV). Our expertise in developing techniques for neutron spectrometry is longstanding, whether it be the calculation of detector responses, development of spectral unfolding techniques, or the determination of a proper protocol for making spectral measurements under difficult field conditions.

Development of a neutron consisting of a single polyethylene cube with passive thermal neutron detectors at varying depths along its major axes will be a step forward from the multisphere spectrometer system, first introduced in 1960 (Bramblett et al., 1960). The cubical spectrometer is a small single unit and therefore will be much easier to

deploy for field measurements than the multisphere system (Sanna and O'Brien, to be published) and will eliminate spatial- and time-dependent effects on the measurements.

The cubical spectrometer consists of a single polyethylene cube, 30 cm on an edge, with TLD-600, TLD-700 detector pairs at various depths along each of the major axes. After incident neutrons are thermalized in the polyethylene material of the cube, they are captured in the TLD-600, which contains almost pure ^6Li having a large thermal capture cross section. TLD-700 contains almost pure ^7Li which has a near-zero capture cross section. The two thermoluminescent detectors otherwise respond identically to other radiations, permitting the TLD-700 detector to be used to evaluate the background of other radiations present in the response of the TLD-600 detector. The symmetrical placement of the detectors in the cube greatly lessens the effect of source anisotropy on the measurement distribution.

Monte Carlo calculations of detector response have been performed for detectors placed along the major axes of the cube. These calculations were performed using the identical combinatorial-geometry Monte Carlo code with which the radiotherapeutic calculations described in 1) above were performed, underscoring the generality and power of these methods. Current work entails the testing of the responses thus obtained, using pseudo-measurement distributions calculated for realistic and idealized single and double spike test spectra. These distributions are being unfolded and the results will be compared to the original spectra to demonstrate that good results can be achieved from measurements made with the cubical spectrometer. The EML Monte Carlo unfolding code SWIFT is being used for these tests (O'Brien and Sanna, 1981; O'Brien and Sanna, 1983).

Future studies include the calculation of the responses for, and testing of, a cube 25 cm on an edge, to see if a significant reduction in weight can be achieved without seriously affecting the spectrometer's ability to unfold neutron spectra from measurement distributions (a 25-cm cube will have only 60% the weight of a 30-cm cube). An experimental study of the isotropy of response of a cubical moderate-and-capture neutron spectrometer is also being planned.

5) Neutron Spectral Unfolding

Neutron spectrometry, as indicated in the section immediately above, is basic to our understanding of the neutron radiation fields to which personnel are exposed. This is especially so due to the poor energy response of neutron personnel monitors. Spectrometry has, however, two constituents: the detector and the analysis that converts the measurements into usable spectra. Neutron spectral unfolding is generally more difficult than gamma-ray spectral unfolding, since the available detectors produce mathematically inconvenient responses. The output of a gamma-ray detector, for instance, bears a strong resemblance to the photon spectrum that caused it.

In order to make use of the newly available Bonner, or multisphere, spectrometer in the mid-1960's, we wrote the first unfolding code applicable to this system, versions of which are still in use at some particle accelerator sites (O'Brien et al., 1965). During the recent study of neutron spectra within reactor containment vessels adverted to several times above, it became apparent that certain tacit assumptions as to the nature of the neutron spectra being measured, which mathematical necessity required to be built into the unfolding codes used by ourselves and others, were responsible in part for differences among the measurements obtained by various groups. To avoid this, we developed a Monte Carlo approach that was free from this weakness and consequently is especially suitable for the study of non-uniqueness and underdeterminedness, two phenomena which make multisphere spectrometry particularly difficult.

Current studies include error propagation in neutron spectral unfolding, the explicit use of initial trial spectra and the application of the Monte Carlo unfolding method to evaluate the performance of the cube spectrometer described above. For the future, much improvement is possible in the mathematical treatment of unfolding, and much remains to be determined. We do not know, (except in the grossest and most empirical way), just how many detectors of what kind are necessary to well represent a given neutron spectrum. Another study of great importance is the question of the accuracy required of the measurements necessary to produce a usable neutron spectrum. Other approaches to Monte Carlo unfolding than the one we currently use are under consideration. These and other studies will be carried on for some time to come.

References

- Bramblett, R. L., R. J. Ewing and T. W. Bonner, A New Type of Neutron Spectrometer, Nucl. Instr. and Meth. 9 (1960)
- Chiu-Tsao, S. T., K. O'Brien and R. Sanna, Dosimetry of Radiation Treatments of Eye Cancer, Trans. Am. Nucl. Soc. 43, 603 (1982)
- Davis, R., Jr., B. T. Cleveland and J. K. Rowley, Underground Science at Homestake, Science Underground, American Institute of Physics, New York
- O'Brien, K., R. S. Sanna and J. E. McLaughlin, Inference of Accelerator Stray Neutron Spectra from Various Measurements, First Symposium on Accelerator Radiation Dosimetry and Experience, Brookhaven National Laboratory, November 3-5, 1965, CONF-651109
- O'Brien, K. and R. Sanna, The Distribution of Absorbed Dose-Rates in Humans from Exposure to Environmental Gamma Rays, Health Physics 30, 71 (1976)
- O'Brien, K. and R. Sanna, The Effect of the Male-Female Body-Size Difference on Absorbed Dose-Rate Distributions in Humans from Natural Gamma Rays, Health Physics 34, 494 (1978)
- O'Brien, K. and R. Sanna, Neutron Spectral Unfolding Using the Monte Carlo Method, Nucl. Instr. and Meth. 185, 277 (1981)
- O'Brien, K. and R. Sanna, Reply to Chambless and Broadway, Nucl. Instr. and Meth. 214, 547 (1983)
- Sanna, R., F. Hajnal and J. E. McLaughlin, Energy Dependence Effects on Neutron Monitor Performance in PWR Containments, Health Physics 43, 263 (1982)
- Sanna, R. and K. O'Brien, A New Type of Moderate-and-Capture Neutron Spectrometer, to be presented at the 6th International Congress of IRPA, Berlin (West), May 1984

RADIOLOGICAL IMPACT OF NUCLEAR FACILITIES

Carl V. Gogolak

Programs are being carried out at EML to determine and quantify potential hazards due to the release of artificial radionuclides from nuclear facilities, both those used in power production and those involved in national defense activities. For this purpose, measurement instrument systems and methods of data interpretation are developed. Field techniques for short-term and long-term monitoring are emphasized, in conjunction with theoretical modelling and laboratory experiments. Program flexibility is maintained to permit a suitable rapid response capability for evaluating specific environmental questions as they arise, and to facilitate cooperative research with other organizations.

The requirement that releases of radioactivity to the environment be kept as low as reasonably achievable has placed the level of incremental exposures from normal plant operations well within the variations of natural background radiation. The direct assessment of environmental radiological effects has thus become increasingly difficult. The currently accepted linear relationship between radiation dose and health effects together with the concept of population dose, however, precludes the acceptability of "not detectable" as a responsible conclusion to monitoring efforts. Theoretical models of the release, environmental dispersion and dose are often relied upon for environmental impact assessments, especially at the design stage. Unfortunately, the errors associated with the use of such models are often ill-defined or unknown.

The EML program on the radiological impact of nuclear facilities addresses specific problems of environmental contamination arising from present or projected nuclear operations. Because of our extensive experience in problems of low level environmental radiation, and our continuing investigations of the pattern and mechanisms of the variations in natural background radiation, we are able to continually improve the lower limits of detection for the incremental exposure to the public. This is achieved through the design of new instruments or modifications of existing ones to improve measurement

performance. However, neither instrumentation in itself nor the mere collection of data is ever sufficient. Careful calibration and quality control procedures are developed to ensure proper performance under adverse field conditions and to meet the exacting standards of precision and accuracy required in this work. Most importantly, new methods of data analysis and interpretation are designed to separate the natural and artificial components of the environmental radiation field. The results of these experimental efforts allow us to test the performance of theoretical dose assessment models, thus increasing the confidence with which they may be used. Currently, we are examining more general statistical procedures which may be applied to environmental monitoring data to increase their usefulness in assessing the consequences of atmospheric pollution, whatever the source.

The main thrust of the Laboratory's research in this field is in developing techniques for environmental measurements of:

- (1) gamma radiation from light-water power reactors with continuously monitoring ionization chambers, solid-state spectrometers and thermoluminescent dosimeters,
- (2) ^{85}Kr beta radiation from nuclear fuel reprocessing plants with thin-wall stainless steel Geiger-Mueller counters, and
- (3) low energy x and gamma radiation from transuranic radionuclides near weapons production or testing facilities with specially-designed proportional counters and thin-window N-type germanium detectors.

In addition to these specific research problems, we continue to maintain and improve our rapid response capability for topical questions of environmental contamination which require attention by DOE. To this end we have computerized the appropriate instrumentation and incorporated them into a mobile laboratory.

The high pressure argon-filled ionization chamber designed at EML for environmental gamma radiation measurement is now commercially available and has come into common use throughout the nuclear industry. The standard deviation analysis developed at EML to separate the exposure rate due to noble gas plumes from natural background variations is based on the difference in the time scales for fluctuations of these two components of the total exposure rate (Gogolak, 1982). We are now investigating time series analysis

of the data in the frequency domain in order to design a suitable moving average digital filter which when applied to the data will remove all slowly varying components of the exposure rate. This should ultimately result in a data processing scheme which is not only more sensitive to small plume exposures, but which is also simpler to use and directly isolates the quantity of interest.

Field gamma-ray spectrometry, initially developed at EML to study fallout and natural background radiation in soils, is now used by many laboratories both here and abroad. The technique is currently being extended to measurements of individual radionuclides in air. In an accident situation at a nuclear facility, the release of radionuclides to the environment may bypass normal effluent monitors so the release rates and radionuclide composition will be in doubt. We have combined our experience in high resolution gamma-ray spectrometry and in atmospheric dispersion modelling to develop a technique for measuring the concentrations and release rates of airborne radionuclides directly and rapidly in the field (Gogolak, 1984). In cooperation with the Institute for Radiation Hygiene of the Federal Health Office of the Republic of Germany, further field tests and refinements of the method are being planned.

EML pioneered the use of thermoluminescence dosimeters (TLD's) for environmental measurements, especially around nuclear facilities. Since these are passive integrating devices the problem of separating natural background from plant contributions requires a different approach from that used with an active continuous monitor. The techniques developed here for this purpose (de Planque, 1979), which permit the assessment of plant related environmental doses as low as 2 mrad per year, have been incorporated in both the American National Standard on environmental use of TLD (ANSI, 1975) and in the corresponding Nuclear Regulatory Commission Guidelines (NRC, 1977) used by all nuclear power facilities in the U. S. Research continues into the effects of various environmental conditions on the response of TLD's.

Reprocessing of spent nuclear fuel for either civilian or military purposes results in substantial releases of the low energy beta emitter ⁸⁵Kr. EML developed a continuously operating environmental detector for this radionuclide with a sensitivity of 3 pCi per liter of air. This was successfully tested in an experiment at the Savannah

River Laboratory (Gogolak et al., 1981). Since noble gases are excellent atmospheric tracers, we are now using the results of this study to further examine the statistical distribution of pollutant concentrations from point source emissions in general, and the accuracy of Gaussian plume dispersion models in particular.

Instances of environmental contamination with transuranic nuclides from weapons production and testing and the potential emergence of civilian breeder reactor technology have highlighted the need for a sensitive environmental detection system for these radionuclides. A gas scintillation proportional counter for laboratory sample analysis (Miller, 1979) and a large area beryllium window proportional counter for field measurements with a sensitivity of 100 nCi per square meter for plutonium, have been developed. Investigations of solid state spectrometers, both coaxial single crystals and planar arrays indicate that the most fruitful area for continuing research will be with N-type germanium, specially packaged for minimum attenuation of low energy radiations.

We plan to continue our efforts to identify potential problems of low level contamination of the environment with artificial radionuclides and to devise appropriate experimental, and analytical procedures to accurately document their levels.

References

- ANSI, Performance, Testing and Procedural Specifications for Thermoluminescence Dosimetry (Environmental Applications), ANSI Report N545-1975, American National Standards Institute, New York (1975)
- de Planque, G., Evaluation of Methods for the Determination of X- and Gamma-Ray Exposure Attributable to a Nuclear Facility Using Environmental TLD Measurements, USNRC Report NUREG/CR-0711 (1979)
- Gogolak, C. V., Rapid Determination of Noble Gas Radionuclide Concentrations in Power Reactor Plumes, Health Physics (in press, 1984)
- Gogolak, C. V., In Situ Methods for Quantifying Gamma-Radiation Levels and Radionuclide Concentrations, IEEE Transactions in Nuclear Science NS-29, 1212-1224 (1982)
- Gogolak, C. V., H. L. Beck and M.M. Pendergast, Calculated and Observed ⁸⁵Kr Concentrations Within 10 km of the Savannah River Plant Chemical Separations Facilities, Atmospheric Environment 15, 497-507 (1981)
- Miller, K. M., A Gas Scintillation Proportional Counter Using a Photomultiplier Tube Array, Nuclear Instruments and Methods 163, 583-586 (1979)
- NRC, Performance, Testing and Procedural Specifications for Thermoluminescence Dosimetry: Environmental Applications, USNRC Regulatory Guide 4.13, Nuclear Regulatory Commission, Washington, DC (1977)

RECONSTRUCTION OF FALLOUT EXPOSURE FROM NEVADA
WEAPONS TESTING

Harold L. Beck

Since early in 1979, EML has been intensely involved in a program designed to reconstruct the radiation exposure to the population of Nevada and surrounding states as a result of nuclear weapons testing at the Nevada Test Site (NTS). Currently about 1200 claims are pending against the U. S. Government alleging injury from such exposure and asking damages amounting to over \$2 billion. The publicity resulting from these claims and the consequent litigation have caused considerable public concern regarding the adequacy of the original post-shot monitoring programs and the credibility of the government's original assurances regarding the safety of the testing program. This controversy has been fueled by a number of studies reported in the scientific literature and in the press alleging increased cancer incidence in the population downwind from NTS as well as by the ongoing controversy regarding the effects of exposure to low level radiation. The importance of reconstructing exposures has been further emphasized by the introduction of several bills in Congress proposing to compensate people who may have been harmed on the basis of the probability of their injury being related to radiation exposure from Nevada testing. Indeed, last January, Congress mandated the Secretary of Health and Human Services to conduct scientific research and prepare analyses necessary to develop valid and credible assessments of the exposure to ¹³¹I to all U. S. citizens from Nevada atmospheric nuclear weapons tests.

In response to the clear need for a careful reassessment of the population exposure to NTS fallout, DOE, in 1979, established a multilaboratory Offsite Radiation Exposure Review Project (ORERP) to estimate doses to the population, beginning with persons living in the limited region covered by the original post-shot monitoring data. Shortly prior to this, at the encouragement of the then Assistant Secretary of Energy for the Environment, and in response to published allegations of increased leukemia incidence in southern Utah as a result of NTS tests, EML began an independent study of fallout exposure in Utah,

most of which is outside the area of post-shot monitoring, and not included in the original ORERP study. The final results of this study were recently reported (Beck and Krey, 1983). Subsequently, while continuing our own assessment, we became special advisors to the ORERP, providing general guidance and assistance in many areas within our realm of expertise. As part of our commitment to the ORERP, we have taken responsibility for the following projects: (1) Reassessing the data for selected U. S. sites from the worldwide fallout monitoring program carried out by EML in the 1950's using gummed-film collectors in order to estimate fallout patterns beyond the region of post-shot monitoring; (2) Serving as technical advisors on a program to study the aerial monitoring data obtained as part of the National Uranium Resources Exploration Program (NURE) for indications of potential NTS fallout "hotspots"; (3) Providing quality assurance support for the ORERP extended regional soil sampling and analysis program.

In addition, EML was recently asked by the Department of Health and Human Services-National Cancer Institute (NCI) to participate in their iodine exposure assessment program as special advisors on fallout deposition.

EML's involvement in these reconstruction studies is consistent with our historical role as one of the foremost centers of fallout research in the world and is considered essential by the sponsors of both the ORERP and NCI studies. In fact, EML was asked to participate in the NCI study even though Congress, in mandating the study, implicitly specified an effort completely separate and independent from the DOE sponsored ORERP project, attesting to EML's reputation for scientific excellence and objectivity.

Aside from our acknowledged expertise in the measurement, assessment, and characterization of environmental radiation fields, EML has generated much of the available data on fallout from both NTS testing, as well as from large scale U. S. and Soviet thermonuclear testing, including data on deposition from the gummed-film and precipitation sampling networks as well as measurements of radioactivity in soil and air. We also developed many of the radiochemical methods used for analyzing trace quantities of fallout nuclides in environmental media.

Most of the methods currently utilized by ORERP to reconstruct fallout exposure rely heavily on information, data, methods and monitoring techniques developed at EML. For

example, the ORERP internal dose estimates are based on relating measured post-shot monitoring data to deposition of various nuclides using theoretical conversions from exposure rate to soil activity developed by EML (Beck, 1980). The current ORERP Phase II extension to areas beyond the post-shot monitoring region is based almost entirely on the unique in situ spectrometric and soil analysis techniques used by EML in assessment of the exposure of the Utah population. This includes precise conformation to EML survey techniques, site selection criteria, soil sampling methods, soil processing methods, and radiochemical analysis methods, all monitored by EML as part of our ORERP quality assurance responsibilities. We expect the NCI iodine exposure study will also rely heavily on EML historical records and samples, particularly our re-analysis of data from the gummed film network.

Another capability developed by EML has been used to obtain an independent corroboration of fallout deposition in Utah. This relies on the analysis of residual long-lived radioactivity in large diameter sediment cores from reservoirs and natural lakes to provide the chronology of fallout deposition. One reservoir near Enterprise in southern Utah was successfully sampled in 1979 (Krey et al., 1980) and several additional lakes and reservoirs were sampled in 1982.

In addition to the research and advisory roles related to our involvement in the reconstruction of NTS fallout exposure, two EML scientists testified for the government in one of the major ongoing litigations, the Allen et al case tried in Federal Court in Salt Lake City. We expect EML staff to be called to testify in other upcoming law suits as well. In addition EML staff members have participated in several briefing sessions for government lawyers as well as in periodic briefings to the Dose Assessment Advisory Group (DAAG), a federal advisory panel charged with overseeing the ORERP.

We expect our involvement in these and related fallout studies to continue for several more years at the current annual level of about 3-4 person years, by personnel from three EML divisions. The ORERP program is expected to continue until at least the fall of 1985 and could be extended. The NCI study is just beginning and is expected to last 3-5 years. Our involvement in upcoming litigation will surely continue for several more years as additional cases come to trial. New legislation could require an even greater effort to document NTS radiation exposures to the entire U. S. population to determine possible compensation.

We expect to concurrently pursue several new areas of research, the need for which has been identified during these studies. Included in these are improvements in our methods for interpreting sediment data, additional studies regarding the correlation between global fallout and precipitation, and improved methods of obtaining representative soil samples in areas where the fallout is not uniformly distributed. The latter topic is of great importance since our soil analysis interpretation presumes the activity in the sample is representative of the actual deposition. A pilot study to test several new and unique sampling methods was carried out this past summer. All of these new research efforts have important applications to the assessment of non-nuclear environmental pollution as well as for radioactivity. In fact, the sediment coring method was developed and is used primarily for the analysis of the temporal history of organic and trace element pollution.

Our efforts in reconstructing the NTS fallout exposure demonstrate the Laboratory's ability to apply our extensive and in many cases unique expertise in the measurement and characterization of low level radiation and radioactivity, developed over many years, to an important national problem, and attests to the wisdom of continuing our core research, development and support efforts designed to maintain and improve our capabilities for measuring and characterizing low-level radiation in the environment.

References

- Beck, H. L., Exposure Rate Conversion Factors for Radionuclides Deposited in the Ground, USDOE Report EML-378 (1980)
- Beck, H. L. and P. W. Krey, Radiation Exposures in Utah from Nevada Tests, Science 220, 18-24 (1983)
- Krey, P. W., E. P. Hardy and M. Heit, Nevada Test Site Fallout in the Area of Enterprise, Utah, USDOE Report EML-372 (1980)

RADIOLOGICAL IMPACT OF ALTERNATE ENERGY SOURCES

Carl V. Gogolak

It is the purpose of the EML study of the radiological impact of alternative energy sources to systematically examine each technology, assess the potential perturbations of the natural radiation environment which may be occasioned by its use, trace those naturally occurring radionuclides which are brought into contact with the biosphere, determine how these radionuclides are redistributed among the combustion or processing products, evaluate the major potential dose pathways, calculate the projected radiological impact wherever possible, and, finally, identify problem areas where further information would be required.

Although there is much research currently focussed on the environmental impacts of alternative energy sources, there has been relatively little which has dealt with radiation problems in a comprehensive manner. Because of our long history of research in environmental radiation, both of variations in natural background levels and of the increments caused by weapons test fallout and the operation of nuclear facilities, our experience in dispersion and dose assessments, and a long standing capability for state of the art low level measurements of natural radioactivity in various media, EML has a natural interest in pursuing these studies.

Issues involving the radiological impact of energy production are no longer confined to nuclear facilities only. An increasing awareness of these problems on the part of the public, together with guidelines for incremental exposures well within the temporal and geographical variations associated with natural background radiation, has focussed attention on the ways in which human activities may affect the radiation environment without necessarily involving the creation of artificial radionuclides. The study of technologically enhanced natural radiation (TENR), has uncovered several areas in which the disturbance and redistribution of radionuclides present in nearly all material can result in radiation exposures to the public which, it has been suggested, may in some cases even exceed those imposed during the normal operation of a light water nuclear power reactor (McBride et al., 1977). Although the scope of TENR is quite wide,

including for example the production of phosphate fertilizer, that portion associated with alternate energy sources is of particular concern to DOE in assessing the overall relative environmental impact of the nation's energy programs. Nearly every energy technology must be considered, for there is little that is undertaken that does not in some way effect human radiation exposure.

Our initial studies of TENR associated with energy production began over a decade ago and resulted in a comprehensive report on the radiological impact of coal combustion in central station electricity generating plants (Beck et al., 1980). This subject continues to draw considerable attention, since in the near term, coal and nuclear are the two sources favored for immediate expansion in the nation's electric power production base (Fabrikant, 1983). Further research in this area is needed, and members of the EML staff have recently participated in a workshop specifically organized to assess the priorities to be assigned to various research issues involved with radionuclides in coal used to generate electric power (Miller and Beck, 1981). We are continuing our measurements of natural radionuclides in coal, bottom ash, fly ash and environmental samples in cooperation with the trace metal and organic pollutant studies being performed as part of EML's non-nuclear research program (Klusek et al., 1983).

The major effort of the present program on the radiological impact of alternative energy sources is now directed towards new, unconventional or emerging energy sources. The priority which is assigned to a given technology depends on several factors:

- (1) the projected importance of its contribution to the nation's total energy supply,
- (2) the time frame in which commercial production facilities are expected to come on line,
- (3) the potential for radiation exposure through its development and use, and
- (4) the extent of existing information, if any, on its radiological impact.

The first of these studies dealt with the potential impact on indoor radon levels resulting from the use of natural gas produced from unconventional sources, including eastern devonian shale deposits, western tight gas sands, geopressurized aquifers, and methane from coal seams (Gogolak, 1980). Most recently we have completed our analysis of the potential environmental exposure to the public from shale oil development (Gogolak, to be published). Currently, we are examining both the gasification and liquefaction methods of coal conversion.

For each technology, a literature review is carried out and potential sources and pathways for natural radionuclides are identified. Our experience in related environmental radiation research allows us to use existing information in new ways, avoiding duplication of effort wherever possible. Prior environmental impact studies are used to help define the source term and potential releases. For example, trace metal analyses which include uranium, potassium and thorium in feedstock and processing wastes have been used for this purpose. Estimates of particulate and fugitive dust releases combined with atmospheric dispersion calculations are also used to aid in estimating the likely increase in local air concentrations of natural radioactivity.

Performing these studies in a systematic way is a great advantage in that information gathered in one study is often applicable to several different problems. Having studied the distribution of natural radionuclides in various coals in assessing the impact of coal-fired power stations, the feedstock concentrations for conversion facilities are more easily defined. Existing information is rarely complete enough for a comprehensive radiological assessment, and we must identify and fill the gaps where necessary. Most often this will involve obtaining a suite of samples for various processes and performing our own analyses. The measurements needed usually include:

- (1) the state of equilibrium of the uranium and thorium series through the process,
- (2) the radon emanating power of the waste products, and
- (3) the fractionation of radionuclides as a function of particle size.

The most challenging aspect of making radiological assessments of this type is in integrating the sparse and sometimes contradictory information on the specific environmental pathways for the radioactivity as the raw materials are processed. For new technologies processing information is usually available primarily from laboratory experiments or pilot plants. Often various competing processes are used to produce a given product. Ideally, from radiological considerations, a model plant whose impact would reasonably well represent that of future production facilities will be formulated in order to make an overall assessment. For some pathways this approach is feasible, in others it is only possible to identify the potentially significant problems and areas where further research is required. It is anticipated that our study of coal conversion will continue

through 1985. Further studies will include advanced oil recovery, geothermal sources (both hot water and hot dry rock), and conservation. The priorities for continued research will be periodically reassessed and will include updated information on technologies already surveyed to incorporate new knowledge gained as they evolve. The ultimate aim of the project remains the development of a complete and systematic survey of the perturbation of the natural radiation environment caused by the development of all new non-nuclear energy technologies.

References

- Beck, H. L., C. V. Gogolak, K. M. Miller and W. M. Lowder, Perturbations of the Natural Radiation Environment due to the Utilization of Coal as an Energy Source, Proc. of the Third International Symposium on The Natural Radiation Environment, USDOE Report CONF-780422, 1521-1558 (1980)
- Fabrikant, I. I., Guest Editorial: Is Nuclear Energy an Unacceptable Hazard to Health? Health Phys. 45, 575-578 (1983)
- Gogolak, C. V., The Radiological Impact of Atmospheric Emissions from Oil Shale Retorts (to be published)
- Gogolak, C. V., Review of Radon-222 in Natural Gas Produced from Unconventional Sources, USDOE Report EML-395 (1980)
- Klusek, C. S., K. M. Miller and M. Heit, Trace Element and Radionuclide Mass Balances at a Coal-Fired Electric Generating Station, Environ. Int. 9, 139-144 (1983)
- McBride, J. P., R. E. Moore, J. P. Witherspoon and R. E. Blanco, Radiological Impact of Airborne Effluents of Coal-Fired and Nuclear Power Plants, USERDA Report ORNL-5315 (1977)
- Miller, K. M. and H. L. Beck, Some Effects of Coal Combustion on the Radiation Environment, Proceedings of the Workshop on Radioactivity Associated with Coal Use, Santa Fe, NM, USDOE Report LA-9106-C (1981)

ENVIRONMENTAL RADIATION

Kevin M. Miller

A major component of human exposure to ionizing radiation comes from natural sources. This "background" radiation has been present throughout the evolution of human life on earth and indeed, since the formation of the earth itself. The advent of nuclear technology in this century has introduced new sources of radiation into the environment and with it, an awareness of the biological effects of radiation, whether it be natural or anthropogenic. The mission of DOE and its predecessor agencies is intimately connected with this technology both with regard to nuclear weapons and commercial uses of nuclear power. The concomitant radiation exposure delivered to workers in these fields as well as the general population has always been carefully studied and in recent years scrutinized to an even higher degree. The assessment of the exposure that results from these sources generally requires a precise knowledge of the natural background component and its spatial and temporal variations because in many cases the magnitude of these variations alone are larger than the contributions from the man-made component under investigation.

EML has had a long history of studying the natural background radiation field both in terms of it being a source of exposure to the population and to differentiate it from any perturbations caused by human activities. In the early 1960's the emphasis was on measuring fallout from atmospheric nuclear weapons tests. In the late 1960's and into the 70's the emphasis shifted to studying the radiation exposure associated with the nuclear fuel cycle, in particular, reprocessing and commercial power plants. In recent years, radioactivity in fossil fuels and the reassessment of fallout from the Nevada Test Site have become principal areas of investigation. In each of these cases, our ability to address the subject matter was predicated on a detailed knowledge acquired over the years from ongoing research of the sources and behavior of natural background radiation. As an example, the reconstruction of the doses delivered to the population of Utah from Nevada weapons test fallout were based on recent measurements of the only major gamma-emitting fission product still present in the soil, ^{137}Cs . Typically, this isotope produces

a mere 3% of the total radiation flux measured in the field, the rest coming from natural radionuclides in the soil. Our ability to isolate and measure this small flux contribution was based on in situ gamma ray spectral techniques developed at EML. This and other projects require development of new instrumentation or the adaptation of state-of-art components for measurement, the collection, handling, and synthesis of data with novel analytical techniques, and perhaps most importantly, interpretation of the results that could only be made with sustained expertise in environmental radiation. The Laboratory's capabilities in present and any future research efforts will depend upon maintaining a program that expands and refines the body of knowledge in this field. The continued interest in and relevance of the subject is best exemplified by the success of three symposia on The Natural Radiation Environment (Adams and Lowder, 1964; Adams et al., 1975; Gesell and Lowder, 1980) that were organized in part by EML as well as two additional special symposia held outside the U. S. (Cullen and Penna Franca, 1977; Vohra et al., 1981).

The uniqueness of EML's capabilities stems from a tradition of applying an appropriate mix of theoretical and experimental investigations into the problem of making low-level radiation measurements in the environment. Pioneering work in applying in situ gamma ray spectrometry using NaI and Ge detectors supported by gamma ray transport calculations has provided a means to determine radionuclide concentrations or inventories in soil as well as their corresponding exposure rates in the air above. The design, development and use of high pressure ionization chambers (HPIC) for environmental radiation studies and the success they have achieved through commercialization of the unit has led to acceptance by industry as one of the primary instruments for continuous monitoring around nuclear facilities. The application of thermoluminescence dosimeters (TLD) for monitoring in conjunction with the development of methods of data interpretation has also been an area of original research. We are continuing in the tradition of adapting or developing the latest technological advances in instrumentation by incorporating into our regimen a more portable spectrometer system, a scintillation survey instrument for accurate exposure rate measurements, and a smaller version of our standard HPIC (Latner et al., 1983). We are also in the process of obtaining a

high resolution combination x and gamma ray detector that will be used for dating the stratigraphic column in lake sediment cores via the analysis of both ^{137}Cs , a man-made fallout product and ^{210}Pb , a natural radionuclide (see material submitted by Heit).

Our present research in environmental radiation from natural sources includes a long term measurement program being carried out at a rural field site at Chester, NJ, about 64 km from EML (Miller et al., 1978-1982). In conjunction with two other field monitoring sites and several employee residences in the New York metropolitan area, temporal variations in the exposure rate associated with the penetrating component of background radiation are being studied (Beck and Miller, 1981; Gulbin and de Planque, 1983). These types of measurements have served as an aid in developing methods for predicting background fluctuations that can be applied to nuclear power plant monitoring (de Planque, 1979, 1980). The data from this network also serves to document any perturbations in the radiation levels that might occur during unusual events such as fallout from a Chinese weapons test (Harley, 1976) or the accident at Three Mile Island Nuclear Power Station (Miller et al., 1979). Our main field site at Chester is also used to test new instruments under realistic conditions in an area where the radiation field is well documented with respect to the source terms and other controlling factors. In addition, short-term specialized experiments that involve the comparison of experimental data with model calculations, as has been done for the radon progeny altitude profile in the atmosphere, are performed on occasion (Beck and Gogolak, 1979).

Another facet of natural background radiation which is currently being studied at the Laboratory is the indoor radiation environment. Since most people in the U. S. spend a high percentage of their time indoors, either in homes or workplaces, it is important to measure the radiation exposure that comes from the natural radioactivity contained in building materials and the effective shielding of outdoor radiation sources provided by the building itself. Further, indoor radiation levels are, to a degree, affected by energy related technologies. The use of flyash or slag from coal combustion in building materials has implications for increasing population exposure as does passive solar heating systems that employ stone-type materials for heat storage. Energy efficient homes which are tightly sealed allow the build-up of radon gas which can

strongly affect internal dose to individuals (see also material submitted by George and Fisenne). Using our knowledge and experience in radiation measurements in the outdoor environment, we are applying similar techniques indoors. These will include screening with TLD's and performing follow-up measurements with gamma-ray spectroscopy and HPIC's. Past work with TLD monitoring in homes and gamma and cosmic-ray transport calculations will also be used as part of our studies. Some of our work in this field has been presented at a recent international symposium (Miller and Beck, 1983).

We expect to continue our background radiation studies into the indefinite future in order to study long term naturally occurring trends and to maintain and improve our capabilities in low-level radiation measurements in general. Indoor radiation studies have just begun and should continue for the next few years until a thorough body of information has been assembled at which point it may become an integral part of our overall commitment to studying the radiation environment.

References

- Adams, J.A.S. and W. M. Lowder, eds. The Natural Radiation Environment, University of Chicago Press (1964)
- Adams, J.A.S., W. M. Lowder and T. F. Gesell, The Natural Radiation Environment II, USERDA Report CONF-720805-P1 (1975)
- Beck, H. L. and K. M. Miller, Temporal Variations of the Natural Radiation Field in Natural Radiation Environment, Proceedings of the Second Special Symposium on Natural Radiation Environment held at Bombay, India, p. 163, Wiley Eastern Ltd. (1981)
- Beck, H. L. and C. V. Gogolak, Time-Dependent Calculations of the Vertical Distribution of ^{222}Rn and its Decay Products in the Atmosphere, J. of Geophys. Res. 84, (1979)
- Cullen, T.L. and E. Penna Franca, eds., International Symposium on Areas of High Natural Radioactivity, Academia Brasileira de Ciencias, Rio de Janeiro, Brazil (1977)
- de Planque, G., TLD Measurements and Model Calculations of Environmental Radiation Exposure Rates, in Proceedings of the Natural Radiation Environment III, USDOE Symposium Series Report CONF-780422-P1, p. 987 (1980)
- de Planque, G., Evaluation of Methods for the Determination of X- and Gamma-Ray Exposure Attributable to a Nuclear Facility Using Environmental TLD Measurements, USDOE Report EML-355 (NUREG/CR-0711) (1979)

Gesell, T. F. and W. M. Lowder, eds., Natural Radiation Environment III, USDOE Report CONF-780422-P1 (1980)

Gulbin, J. and G. de Planque, Ten Years of Residential TLD Monitoring, Presented at 7th International Conference on Solid State Dosimetry, Ottawa, Canada (1983) (proceedings to be published)

Harley, J.H., HASL Measurements of Fallout Following the September 26, 1976 Chinese Nuclear Test, USERDA Report HASL-314 (1976)

Latner, N., K. Miller, S. Watnick and R. T. Graveson, SPICER: A Sensitive Radiation Survey Instrument, Health Phys. 44, 379-386 (1983)

Miller, K. M. and H. L. Beck, Indoor Gamma and Cosmic Ray Exposure Rate Measurements Using a Ge Spectrometer and Pressurized Ionization Chamber, Presented at International Seminar on Indoor Exposure to Natural Radiation and Related Risk Assessment, Anacapri, Italy (1983) (proceedings to be published)

Miller, K. M., G. de Planque, J. F. Gulbin and M. C. Boyle, Environmental Radiation Monitoring in Regional Baseline Station, Chester, N.J., USDOE Reports EML-347, EML-367, EML-383, EML-399, EML-411, issued annually 1978, 1979, 1980, 1981, 1982

Miller, K., C. Gogolak, M. Boyle and J. Gulbin, Radiation Measurements Following The Three Mile Island Reactor Accident, USDOE Report EML-357 (1979)

Vohra, K. G., U. C. Mishra, K. C. Pillai and S. Sadasivan, eds., Natural Radiation Environment, Wiley Eastern Ltd., Bombay, India (1981)

NATURAL RADIOACTIVITY IN THE ATMOSPHERE

Herbert W. Feely

The ongoing operation of the EML atmospheric sampling programs (see submissions of Hardy et al. and Leifer) has provided the opportunity to analyze samples for natural radionuclides for special projects covering a wide variety of interests.

One such project is the study of atmospheric circulation. The group of natural radionuclides that are the products of the bombardment of the atmosphere by cosmic rays have at times been used as tracers of the circulation of the atmosphere. Filter samples of air are collected for radiometric and radiochemical analysis during both the EML program of stratospheric sampling, the High Altitude Sampling Program (HASP), and the program of sampling ground level air, the Surface Air Sampling Program (SASP). While these filters are measured routinely for several fission products by means of gamma spectrometry, the spectra from these measurements contain peaks attributable to several natural radionuclides as well, and those for the cosmic ray product, beryllium-7, are normally large enough to permit the accurate calculation of the concentration of this nuclide in the air. Consequently concentrations of this nuclide are measured and reported regularly by these programs. The mean concentrations of beryllium-7 and the seasonal variations in these concentrations at the surface air sampling sites have been observed to reflect regional differences in atmospheric circulation and precipitation rates at those sites (Feely et al., 1979). Thus the mean concentrations of beryllium-7 at Barrow, Alaska and at other Arctic sites are lower than the mean concentrations at temperate sites, even though stratospheric concentrations of beryllium-7 reach their maximum above the polar regions. This indicates that the stability of the Arctic troposphere strongly inhibits the vertical transport of air within that atmospheric region. The maximum beryllium-7 concentrations at Barrow occur during the winter months, when air from mid-latitudes carries into the Arctic both higher concentrations of this nuclide and high concentrations of the pollutants which produce the "Arctic haze". We continue to study beryllium-7 concentrations at all of our SASP sites because it serves as a tracer for such atmospheric processes.

In another special project some interesting results have been obtained from measurements of radon and its daughter products, lead-210 and polonium-210, in samples collected as part of one HASP mission, and from measurements of lead-210 in samples collected at several SASP sites. In May 1930, following the eruption of Mount St. Helens, it was reported that very high concentrations of radon had been measured in the tropospheric plume from the eruption (Fruchter et al., 1980). The stratospheric plume from that eruption was intercepted by a HASP flight two days after the eruption, and radon was analyzed in gas samples and lead-210 and polonium-210 were analyzed in filter samples collected on that flight (Leifer et al., 1981). It was found that the eruption had not injected detectable amounts of these radionuclides into the stratosphere.

Lead-210 has been measured at times in filters from the SASP (Feely and Sanderson, 1981). It was observed that lead-210 concentrations reach a seasonal peak in the Arctic during the winter months, when the polluted air masses which produce the Arctic haze layers are reaching that region from the Asian continent. At Mauna Loa Observatory the lead-210 concentrations reach a seasonal peak during the Spring at the same time that dust which originates in Asian dust storms is being carried across the Pacific and past those islands. Thus it appears that lead-210 is a possible tracer of air which has been in contact with the deserts of Central Asia. We are continuing to measure lead-210 in filters from several sites, including Barrow and Mauna Loa, to determine whether it provides information on atmospheric circulation which is not otherwise available.

References

- Feely, H. W., H. L. Volchok and C. G. Sanderson, Seasonal Variations in the Concentration of ^7Be in Surface Air (abstract), in The Natural Radioactive Isotopes of Beryllium in the Environment, A Symposium held at Yale University (1979)
- Feely, H. W. and C. G. Sanderson, Atmospheric ^{210}Pb at Barrow Observatory and Mauna Loa Observatory, in Geophysical Monitoring for Climatic Change, Summary Report (1981)
- Fruchter, J. S., et al., Mount St. Helens Ash from the May 18, 1980 Eruption: Chemical, Physical, Mineralogical, and Biological Properties, Science 209, 1116-1125 (1980)
- Leifer, R., L. Hinchliffe, I. Fisenne, H. Franklin, E. Knutson, M. Olden, W. Sedlacek, E. Mroz and T. Cahill, Measurements of the Stratospheric Plume from the Mount St. Helens Eruption: Radioactivity and Chemical Composition, Science 214, 904-907 (1981)

RADON PROGRAMS

Andreas C. George

Introduction

Because of the world energy situation efforts have been made to reduce energy consumption or seek alternative fuels. Since more than one-third of U. S. energy is consumed in the heating and cooling of buildings, this area is a prime target for reduction efforts. A survey in 1980 showed that in this effort 40% of home owners added insulation to their houses. This emergence of energy conservation is causing the quality of indoor air to grow worse (NRC, 1981). Since people in the U. S. on the average spend between 75 to 80% of their time indoors there is a potential for increase in the exposure of the population to a number of harmful pollutants. Among the more important indoor air pollutants are tobacco smoke, carbon monoxide, nitrogen dioxide, formaldehyde and radon plus its daughters (NRC, 1981).

Indoor air pollution as a national issue was recognized in the last 10 years when research results suggested that indoor exposures to several pollutants may be significant. Interest in indoor air quality and recognition of a need for substantial and continuing research has been growing at a fast pace. Coverage by the media as well as research activity expressed in scientific conferences is increasing. Most recent examples are: "Workshop on Indoor Air Quality Research Needs" (EPA, 1980) sponsored by the Department of Energy, Assistant Secretary for Conservation and Renewable Energy and by the United States Environmental Protection Agency, Office of Environmental Engineering and Technology, 1981; testimony before the House Science and Technology Joint Subcommittees on the "Threat from Indoor Air Pollution", August 2 and 3, 1983; "International Seminar on Indoor Exposure to Natural Radiation and Related Risk Assessment" held in Capri, Italy, October 3-5, 1983; and an ASHRAE/DOE sponsored conference in Santa Barbara, October 10-21, 1983, on "Management of Atmospheres in Tightly Enclosed Spaces".

In spite of the accumulating scientific evidence, the problem of indoor air pollution has been largely ignored (Kirch, 1982). Government agencies are now beginning to recognize the responsibility to evaluate the possible health and environmental affects

associated with energy conservation programs and inform the public of the possible consequences on the indoor environment.

As mentioned above one of the major pollutants in the indoor environment is radon and its daughters. They contribute a significant portion of the background radiation dosage in the environment. Investigations during the past few years have revealed that inhalation of radon and its daughters can result in radiation doses which exceed by far all others from natural sources of ionizing radiation (NRC, 1981). The exposure of people to radon and its daughters is poorly documented. Information to derive a population distribution is inadequate and the present estimate of the average human exposure is uncertain. Therefore there is an urgent need to obtain more meaningful data that will provide the desired basis of comparison with other sources of radiation exposure.

Our present Radon Program evolved from EML's field oriented work (1950-1972) which was directed to the measurement and reduction of occupational exposure to uranium and decay products in uranium mines, concentration mills and in nuclear fuel manufacturing facilities. In the last 8 years, EML has turned to non-occupational environments. The main thrust of this effort was to characterize indoor radon and daughters in several regions of the U. S. EML developed several instruments for this purpose such as the charcoal canister for measuring radon flux (Countess, 1976) the modified Marinelli collector (Countess, 1978) for measuring radon in water, the PERM (George and Breslin, 1977) and the activated carbon device (George, 1983) for measuring radon concentration in air and the MOD (Guggenheim et al., 1979) and the integrating working level monitor (Latner et al., 1981) for measuring radon daughter concentrations in air. All these instruments were designed, built and evaluated at EML and many have been copied by commercial instrument companies for field applications.

While the concentrations of indoor air pollutants is the most important parameter to measure, characterization of the aerosol and ventilation measurements is also an integral part of the overall program providing specific information about the properties of the air contaminants, particle size, source distribution in structures and respiratory deposition.

EML's radon calibration facilities and aerosol technology provide the necessary environment in which all the aforementioned instruments and techniques can be evaluated. Through the sponsored radon intercomparison exercises, and workshops (Fisenne et al., 1981; 1983; USDOE, 1983), EML provides the necessary quality assurance needed by the participating groups involved with radon programs (see submission by Fisenne).

Following are specific programs we intend to pursue in the foreseeable future. These are selected to advance the state of the art, where present techniques are limited, and gather data to enable us to assess the risk from energy related sources and technology.

1) EML Contributions to a National Survey for Radon

This is badly needed to establish the distribution of exposures and allow adequate estimates of the average population exposure. It will also identify that portion of the population exposed to excessive concentrations of radon and its daughters. Results from regional studies conducted by EML (George and Breslin, 1980; George and Eng, 1983; George et al., 1983; Fleischer et al., 1983; Alter and Oswald, 1983), indicate that a substantial fraction of homes measured have radon levels exceeding the U. S. Nuclear Regulatory Commission's limit of 3 pCi/l for continuous exposure in uncontrolled areas.

Because EML's funding for personnel and equipment is limited, we cannot undertake a national survey for indoor radon. However, EML will be available to assume a significant role in the design of such a survey. The experience gained by EML while conducting comprehensive studies of indoor radon in about 100 buildings (George and Breslin, 1980; George and Eng, 1983; George et al., 1983; George et al., 1983) can help plan the proposed broader surveys. Moreover, the traditional role of EML as a troubleshooting laboratory can serve the interests of DOE by overseeing the quality of the work of others in addressing the risk from energy related programs. In the interim, EML will continue to design instruments and improve the state of the art while conducting laboratory research and small scale surveys.

2) Measurements of Physical Properties of Radon Daughters

To accurately assess the health effects of radon exposure, the physical properties of its daughters and the environmental factors that influence them must be known. Although there has been considerable recent progress (Sinclair et al., 1977; George and Breslin, 1980) a more substantial research effort is needed to study the following phenomena.

- a) To study size distribution of radon daughter aerosol with specific emphasis on the unattached fraction of radon daughters that is thought to contribute most of the dose to the respiratory tract.
- b) To determine the nature, rate and extent of attachment of radon daughters to aerosol particles.

- c) To measure removal rate of radon daughters to surfaces by filtration or by plateout (George et al., 1983; Knutson et al., 1983) mechanisms. Research in this area may contribute to indoor radon daughter control.

Since EML has already developed the instrumentation required to characterize most of the above parameters, we plan to measure them in a sample of buildings representative of U. S. housing. We will continue to improve the instruments and methods for a better estimate of the physical characteristics of radon daughters. We also foresee a need for an international intercomparison on particle size measurements of radon daughters, since results obtained in different parts of the world appear to be discordant.

3) Development and Refinement of Thoron Measurement Methods

There is a need to determine the thoron contribution to total dose from radon and thoron daughters. To accomplish this, accurate and practical measurement techniques for thoron and its daughters must be developed. There is limited evidence that the contribution of thoron to the total working level can be significant under some circumstances, one of which is likely to be high thorium content in building materials.

EML's radon chamber can be easily adapted to undertake this assignment. We will generate known atmospheres of radon-thoron to test the measurement techniques that will be used to determine the contribution of thoron daughters. We expect to sample a number of buildings in New York, New Jersey and Pennsylvania for this purpose.

4) Sources of Indoor Radon

Research is needed to define the sources of indoor radon and the mechanism of delivery into structures. The factors that influence radon concentrations in structures such as the radium content of soil and building materials, radon flux and pathways of radon entry into structures should be known. Information of this type might lead to the development of standards that will point out sites that are indicative of the potential for elevated radon exposures.

To fully understand the impact of radon indoors, we will measure radon sources, ventilation rates and air concentrations in the number of homes and calculate material balance.

References

- Alter, W. H. and R. A. Oswald, Results of Indoor Radon Measurements Using the Track Etch Method, Health Phys. 45, 425 (1983)
- Countess, R. J., Radon-222 Flux Measurements with Charcoal Canister, Health Phys. 31, 456 (1976)
- Countess, R. J., Measurement of Radon-222 in Water, Health Phys. 34, 390 (1978)
- Environmental Protection Agency, Workshop on Indoor Air Quality Research Needs, CONF-801259, EPA Report 600/7-81-118 (1980)
- Fisenne, I. M., A. C. George and M. McGahan, Radon Measurement Intercomparisons at EML, USDOE Report EML-397 (1981)
- Fisenne, I. M., A. C. George and H. W. Keller, The 1982 Radon Intercomparison Exercises at EML, USDOE Report EML-413 (1983)
- Fleischer, R. L., A. Magro-Campero and L. G. Turner, Indoor Radon Levels in the Northeastern U.S.: Effects of Energy Efficiency in Homes, Health Phys. 45, 407 (1983)
- George, A. C. and A. J. Breslin, Measurement of Environmental with Integrating Instruments, Workshop on Methods for Measuring Radiation in and Around Uranium Mills, E. P. Harward, Ed., Atomic Industrial Forum, Inc. 3 (1977)
- George, A. C. and A. J. Breslin, The Distribution of Ambient Radon and Radon Daughters in Residential Buildings in the New Jersey-New York Area, in Natural Radiation Environment III, Vol. 2, CONF-780422 (1980)
- George, A. C. and J. Eng, Indoor Radon Measurements in New Jersey, New York and Pennsylvania, Health Phys. 45, 397 (1983)
- George, A. C., E. O. Knutson and H. Franklin, Radon Measurements in Solar Buildings Health Phys. 45, 413 (1983)
- George, A. C., E. O. Knutson and K. W. Tu, Radon Daughter Plateout - I Measurements, Health Phys. 45, 439 (1983)
- George, A. C., M. Duncan and H. Franklin, Measurements of Radon in Residential Buildings in Maryland and Pennsylvania, Presented at the International Seminar on Indoor Exposure to Natural Radiation and Related Risk Assessment, Capri, Italy, October 3-5, 1983

- George, A. C., Integrated Measurement of Indoor Radon Using Activated Carbon, Health Phys. (in press, 1983)
- Guggenheim, F. S., A. C. George, R. T. Graveson and A. J. Breslin, A Time Integrating Environmental Radon Daughter Monitor, Health Phys. 36, 452 (1979)
- Kirsch, L. S., Behind Closed Doors: Indoor Air Pollution and Government Policy, The Harvard Environmental Law Review, Vol. 6 (1982)
- Knutson, E. O., A. C. George, J. J. Frey and B. R. Koh, Radon Daughter Plateout-II, Prediction Model, Health Phys. 45, 445 (1983)
- Latner, N., S. Watnick and R. T. Graveson, Integrating Working Level Monitor EML Type TF-11, USDOE Report EML-389 (1981)
- National Research Council, Indoor Air Pollutants, Committee on Indoor Pollutants, National Academy Press, Washington, DC (1981)
- Sinclair, D., A. C. George and E. O. Knutson, Application of Diffusion Batteries to Measurement of Submicron Radioactive Aerosols, American Nuclear Society National Meeting Papers, ANS Meeting, San Francisco, CA (1977)
- U. S. Department of Energy Report EML-416, EML Indoor Radon Workshop, A. C. George, W. Lowder, I. Fisenne, E. O. Knutson and L. Hinchliffe, Editors, New York (1983)

ATMOSPHERIC TRACER STUDIES

Raymond J. Lagomarsino

Increasing concern by government, industry and the public over the international, national and regional aspects of air pollution from power plants and industrial sources, under both routine operational and accident conditions, has created a need for reliable model simulations of transport and dispersion as far as 1000 kilometers from the source. Experimental verification of transport and dispersion calculations is essential to determine the validity of the models, environmental assessments, based on model simulations, and the proposed fuel or pollution control strategies to reduce the impacts of "acid rain".

Verification of model calculations using actual radioactive and non-radioactive pollutants is difficult because of the multiplicity of sources and imprecise knowledge of actual emissions. It therefore is generally recognized that non-reactive, non-depositing tracers which can be released at precisely controlled rates and locations are needed. The ideal tracer material must be non-toxic, should have an atmospheric residence time of at least several weeks and there should be no other source either natural or anthropogenic. Since the atmospheric dilutions are very large, the tracer material must be measurable in very low concentrations. Based on these requirements, several candidate tracers were suggested (Lovelock, 1982) from which two perfluorocarbons, perfluoromethylcyclohexane and perfluorodimethylcyclohexane were selected and appropriate tracer release and analysis systems developed and tested.

Because of extensive experience in environmental sampling and field testing and demonstrated expertise in high quality, ultra low-level gas chromatography-electron capture analysis developed for the High Altitude Sampling Program, EML's participation in the tracer program was a natural, highly productive and cost-effective utilization of resources.

EML has been responsible for the collection and analysis phases of several DOE-NOAA atmospheric tracing projects. These projects include a successful test and demonstration of the long range atmospheric tracer system in 1981 (Ferber, 1982), and two separate

experiments in the geothermal areas of northern California 1981-1982 as part of DOE's Atmospheric Studies in Complex Terrain (ASCOT) (Gudiksen, 1982, 1983), which was designed to determine the impact of hydrogen sulfide emissions from present and planned geothermal power plant cooling towers during nocturnal drainage winds.

The application of this technology to general and specific concerns of the DOE are broad and varied. At the present time the Cross Appalachian Tracer Experiments (CAPTEX), which are a joint US-Canadian venture, are viewed as the first of a series of tracer studies of long range transport and dispersion across the Appalachian Mountains from the Ohio Valley and Sudbury, Ontario. CAPTEX should prepare the way for greater effort to follow, both in the DOE-NOAA program and in the private sector. The program is important in the acid rain controversy, which continues to be an environmental problem with both national and international implications. The CAPTEX is designed to lead to the development and validation of models which will presumably help resolve some of the controversial aspects of the acid rain situation and permit the assessment of various strategies to reduce the adverse affects. The Electric Power Research Institute (EPRI), the Environmental Protection Agency, the Atmospheric Environmental Service of Canada (AES) and the Ministries of Environment of Ontario and Quebec are actively participating in the program.

In another effort, EML will participate with NOAA, LLL and BNL in the ASCOT program developed to identify and understand the effects of local terrain on pollutant dispersion. This program is significant because most experimental data on atmospheric transport have been obtained in areas of relatively flat terrain, while in fact many fossil fuel reserves and power production facilities are located in complex terrain.

A series of experiments are planned in Colorado during 1984 in which perfluorocarbons will be used to study valley flows and larger scale flows in anticipation of atmospheric pollution concerns attendant to oil shale development.

The transport and dispersion of pollutants over an urban area is another DOE concern because of the high concentration of pollutant sources in and around cities. To allow model development and validation, EML will participate in a year long dispersion

experiment (METREX) sponsored by DOE-NOAA which will be conducted in Washington, DC. The experiment, scheduled to start in November 1983, will involve the release of two different tracers from roof tops at two different locations. Automatic sequential samplers will be utilized at three sites to collect 8 hour samples throughout the year.

The Electric Power Research Institute (EPRI) is exploring the concept of a Massive Aerometric Tracer Experiment (MATREX) that may involve a year long series of multiple tracer releases with samplers covering the eastern U. S. The experiment in which EML will also participate, is designed to study the long range transport and deposition of acid rain precursors and attempt to establish quantitative relationships between emissions and acid deposition. This experiment may begin in 1986.

It appears, therefore, that the Laboratory will continue to be involved in atmospheric tracer programs, at approximately the current level of effort, continuously, for at least the next 3 or 4 years.

References

- Ferber, G., K. Telegadas, J. Heffter, C. Dickson, R. Dietz and P. Krey, Demonstration of a Long-Range Atmospheric Tracer System, Interagency Energy/Environment RLD, Program Report, EPA-600/7-81-006 (1981)
- Gudiksen, Paul H., Ed., ASCOT Data from the 1981 Cooling Tower Plume Experiments in the Geysers Geothermal Area, UCID-19332, II, Section 7-49 (1982)
- Gudiksen, Paul H., Ed., ASCOT Data from the 1980 Field Measurement Program in the Anderson Creek Valley, California, UCID-18874-80, III, pp. 1202-1316 (1983)
- Lovelock, J. E., G. J. Ferber, Atmospheric Environment, 16, 1467-1471 (1982)

AEROSOL PROGRAMS

Earl O. Knutson

Introduction

EML's aerosol programs are driven by two major environmental concerns, both of which emerge as a result of energy options in the U. S. One relates to the possible ecological affects of the release of acid gases into the atmosphere by the burning of fossil fuels, particularly high sulfur coal, and the other stems from the possible health effects due to conservation. The gases released, especially oxides of sulfur and nitrogen, act on trees, lakes and soils by wet and dry deposition, either directly or after reacting in the atmosphere to form aerosol particles. "Acid rain" is a very useful descriptive term for this phenomenon, provided it is understood to include dry as well as wet deposition (see submission by Feely and Bogen). Acid rain is recognized as a matter for concern by both the public and the scientific community (NRC, 1983), with some responsible scientists advocating action beyond research. As a result, EML is continuing and gradually expanding the research effort in this area.

Energy conservation measures, such as reducing the fresh air intake of buildings, may affect future health by sealing inside those pollutants which have prominent indoor sources. Even apart from conservation, there is evidence that indoor pollution is a matter for some concern (NRC, 1981). It is conceivable that conservation will change the indoor air quality sufficiently to bring about measurable health effects which may suggest foreclosure of the conservation option.

EML's aerosol programs attack carefully selected facets of the above concerns. In the ecological area, our main work is to characterize the rural aerosol which is one of the avenues for potential insult to trees, lakes and soils in the northeast U. S. EML's Regional Baseline Station at Chester, NJ, is a near-ideal location for collecting samples, which are analyzed for concentration, particle size distribution and chemical composition. Using the results of sampling, aerosol physics calculations are applied to help understand the deposition process. There is a close relationship between the aerosol programs and the deposition programs, discussed separately (see submission by Feely and Bogen).

With regard to indoor air quality, we seek to determine the properties of the indoor aerosol by measurements either in houses or in the laboratory, using common sources of indoor aerosol. Indoor aerosols are an important component of indoor air quality because they may contain non-nuclear chemical irritants as well as radioactivity. Our work on indoor aerosols is closely related to EML work on indoor radon and radon daughters, which is also discussed separately under natural radioactivity (see submission of George).

Current and Recent Work on Aerosol Programs

Work at EML's Regional Baseline Station near Chester, NJ, has consisted of a series of sampling/measurement projects to determine the particle size and other characteristics of the typical rural Northeast aerosols. Work done in 1980 on the particle size distribution of Pb and ^{210}Pb (Knuth et al., 1983) has convinced us that if aerosol dry deposition is ever to be understood and modelled, there must first be accurate and reliable particle size data on the rural atmospheric aerosol. Recently, in the autumn of 1983, the sampling schedule was greatly accelerated during the Cross Appalachian Tracer Experiment (CAPTEX) to take advantage of sampling in air masses of known origin and history (see submission of Lagomarsino).

In earlier work at Chester, an automated diffusion battery and condensation nucleus counter was run for more than two months in autumn of 1979. This apparatus, developed at EML as part of the aerosol program, is notable for its sensitivity to particles smaller than $0.01\text{ }\mu\text{m}$, such as particles formed by photochemical conversion of power plant effluent gases. It was found that the size distributions obtained could be interpreted in terms of the modern "trimodal" theory of atmospheric aerosols. Included in the data were several cases of pure nucleus mode aerosol (Knutson, 1980).

Also in progress is a study using electron microscopy to characterize aerosol samples taken at Chester. One specific objective of this study is to determine if there is any difference in kind between samples collected during high SO_4 atmospheric conditions and those taken in normal conditions. The broader objective is to characterize rural Northeast aerosols, watching particularly for such energy-related substances as coal and oil fly ash. Also of great interest is the morphology and other features of sulfate-bearing particles, which probably trace to coal burning power plants.

With regard to indoor aerosols, we have work underway to determine the sources of indoor aerosols and to characterize particles from common sources. In a brief, preliminary study, electron microscopy/x-ray fluorescence was used to study filter samples of aerosol taken in residences of several EML employees. The main finding of this work, not surprising once the results were in, was that gas cooking is the main source of aerosol in residences. Some houses with attached garages also showed traces of auto exhaust particles. This was a pilot study which showed that, with optimal sample collection, electron microscopy and x-ray fluorescence can be very useful in identifying sources of indoor aerosols.

A laboratory study was done to determine the amount and characteristics of particles from small portable space heaters, electric or kerosene-fired (Tu and Hinchliffe, 1983). These heaters have become popular because they use fuel efficiently, heating only the area desired. Emission factors were developed, and the aerosol from the kerosene heater was checked for polycyclic aromatic hydrocarbons (no significant amount was found).

This study of particle emissions from space heaters was followed up by a study of the total respiratory deposition of submicrometer particles, including those formed by kerosene heaters (Tu and Knutson, 1983). EML was in a position to do this needed study because of our expertise in working with submicrometer aerosols.

Another investigation of the sources of indoor aerosol was done using an automated laser-illuminated single particle optical counter, programmed to sample each hour from both indoors and outdoors. The results confirmed that in a well-ventilated structure the indoor and outdoor aerosol concentrations strongly correlated, showing that the indoor aerosol comes from outdoors. Conversely, in a poorly ventilated building indoor aerosol correlated with indoor activity rather than the outdoor aerosol, showing that the sources are indoors.

Laboratory research is also conducted to improve on aerosol measurements through a study of the basic physics of aerosols. The goal is to look beyond the most pressing current issues and to provide the means for better measurements in the future, even if the issues and questions are not as yet completely in focus.

Much of our recent laboratory work in aerosol studies has dealt with condensation nucleus counters and other methods of measuring the number concentration of sub-micrometer aerosols, particularly with such questions as the reliability, accuracy, precision and comparability of these measurements (Knutson et al., 1982; Sinclair, 1982; Sinclair, 1983a; Sinclair, 1983b; Sinclair and Yue, 1982; Yue, 1982). Work has been done also on inertia-based size selective samplers for supermicrometer particles (Knutson, 1980; Knuth, 1983) and on a commercially available but largely unknown ultra high volume electrostatic aerosol sampler (Tu, 1983).

Custom-generated aerosols with known composition and particle size are needed for accurate calibration of instruments, as well as other aerosol physics experiments. Our recent work in this area has stressed environmentally significant materials such as ammonium bisulfate and pyrene (Tu, 1982; Tu, 1983).

Future Work

With regard to the northeast U. S. regional aerosol, we plan to continue size-selective sampling at Chester for a period of about three years. There may also be intensive, campaign-type studies similar to CAPTEX.

With regard to indoor aerosols, our recent work on sampling in residences has proven very useful. We plan to continue this on a research basis, improving our methods where possible, and to continue laboratory experiments to characterize aerosol emissions for household appliances.

Work done in 1970's on low pressure impaction led to sampling and analysis techniques that have been a key part of EML's stratospheric sampling program. However, the basic physics and chemistry of the process are still not very well understood. Future efforts will be directed in this area.

In addition, we intend to concentrate on methods of generating and doing experiments with aerosol particles of diameter below 10 nm. This is needed to better calibrate aerosol samplers used in studies of unattached radon daughters and of the "nucleus mode" of the atmospheric aerosol.

References

- Knuth, R. H., E. O. Knutson, H. Feely and H. L. Volchok, Size Distribution of Atmospheric Lead and Lead-210 in Rural New Jersey: Implications for Wet and Dry Deposition, in, Precipitation Scavenging, Dry Deposition and Resuspension, Coordinated by H. R. Pruppacher, R. G. Semonin and W.G.N. Slinn, Elsevier, pp 1325-1335 (1983)
- Knuth, R. H., Calibration and Field Application of a Sierra Model 235 Cascade Impactor, Am. Ind. Hygiene Assoc. J. (in press 1983)
- Knutson, E. O., Assessment of a Modified Cascade Centripeter, Am. Ind. Hyg. Assoc. J. 41, 185-190 (1980)
- Knutson, E. O., Aitken Particle Number Concentration and Size Distribution, USDOE Report EML-383, pp. 37-58 (1980)
- Knutson, E. O., D. Sinclair, K. W. Tu, L. E. Hinchliffe and H. Franklin, An Inter-comparison of Aerosol Instruments - Number Concentration, USDOE Report EML-403 (1982)
- National Research Council, Indoor Pollution, National Academy Press, Washington, DC (1981)
- National Research Council, Acid Deposition - Atmospheric Processes in Eastern North America, National Academy Press, Washington, DC (1983)
- Sinclair, D., Particle Size Sensitivity of Condensation Nucleus Counters, Atmos. Envir. 16, 955-958 (1982)
- Sinclair, D., Intrinsic Calibration of the Pollak Counter - a Revision, Paper in preparation (1983a)
- Sinclair, D., Measurements of Temperature and Drop Size in the Pollak Condensation Nucleus Counter, Paper in preparation (1983b)
- Sinclair, D. and P. C. Yue, The Continuous Flow Condensation Nucleus Counter, II, Aerosol Sci. Tech. 1, 217-223 (1982)
- Tu, K. W., A Condensation Aerosol Generator System for Monodisperse Aerosols of Different Physiocochemical Properties, J. Aerosol Sci. 13, 363-371 (1982)

Tu, K. W., An Evaluation of an Ultra High Volume Airborne Particle Sampler, the LEAP, American Ind. Hyg. Assoc. J., (in press 1983)

Tu, K. W. and L. E. Hinchliffe, A Study of Particulate Emissions from Portable Space Heaters, American Ind. Hyg. Assoc. J., (in press 1983)

Tu, K. W., Monodisperse Aerosols of Ammonium Bisulfate and Pyrene, American Ind. Hyg. Assoc. J. (in press, 1983)

Tu, K. W. and E. O. Knutson, Total Respiratory Deposition of Hygroscopic and Hydrophilic Submicrometer Aerosols, Paper in preparation (1983)

Yue, P. C., Comparison of Two Identical Continuous Flow Nucleus Counters, Atmos. Environment 16, 2745-2747 (1982)

POLLUTANTS IN THE UPPER ATMOSPHERE

Robert Leifer

Introduction

The High Altitude Sampling Program (HASP) is composed of Projects Airstream (aircraft sampling) and Ashcan (balloon sampling) and provides measurements of radioactivity and non-nuclear compounds in the upper troposphere and stratosphere. For many years the major driving force for HASP was the investigation of radioactivity in the stratosphere with emphasis on maintaining inventories of ^{90}Sr originating from atmospheric nuclear tests and needed by researchers to evaluate human population exposure. A description of this part of the program can be found in the material submitted by Hardy et al. on "The Global Fallout Program".

While the original purpose of this program was the measurement of radioactivity in the stratosphere it was later realized that HASP had an inherent non-nuclear capability. The impetus to measure trace gases and aerosols, as well as radionuclides, was based on the need to evaluate the impact of increasing emissions of anthropogenic pollutants on human health and the earth's climate. The advantages of simultaneous measurement of stable compounds and radioactivity in atmospheric samples has been extremely valuable to EML. For example, because of this capability we were able to estimate the concentrations of radon, ^{210}Pb , ^{210}Po , and aerosols (sulfuric acid and ash content) in the plume from the eruption of Mt. St. Helens in 1980 and eliminate concern about elevated radon concentrations on public health (Leifer et al., 1981). No other research group in the world has this dual capability.

Freons and the Ozone Problem

In a cooperative and simultaneous effort with Los Alamos, NASA, Johnson Space Center, and Environment of Canada we measure sulfate, nitrate, and ammonium ions, aerosol size and mass distribution, individual particle composition, composition of bulk impactor samples and more than 20 trace gases including halocarbons, CO , CO_2 , CH_4 , CO_s , column ozone and column SO_2 . Our involvement in trace gas measurements dates back to the early 1970's when Dr. Lester Machta of NOAA suggested that CCl_3F

would be a good tracer of atmospheric mixing and circulation. Subsequently Molina and Rowland (1974) showed that the Freons interacted with ozone, and could lead to an increase in the ultra violet radiation on the ground, possibly increasing the risk of skin cancer.

Because of DOE's regularly scheduled flights in the stratosphere and our already established capability for measuring CCl_3F and N_2O we were able to expand and improve the instrumentation for collection and analysis of trace gases on the aircraft. We are presently measuring CCl_3F , CCl_2F_2 , CH_3CCl_3 , CCl_4 , and N_2O , compounds directly related to the ozone problem.

Since 1974, we have measured CCl_3F in the stratosphere, provided data on 2-dimensional distributions, inventories and trends, and calculated lifetimes of this halogen. These data represent the longest continuous measurement record of CCl_3F in the stratosphere. New compounds, related to the ozone problem such as CH_3CCl_3 , have been added to the program as they are identified by the scientific community. These measurements are continuing to be made because they are needed as inputs into multidimensional transport models.

Sulfur in the Stratosphere

Sulfur compounds, both anthropogenic and natural, emitted at the earth's surface interact with both long and short wave radiation from the earth and sun respectively. Sulfur dioxide, released to the atmosphere during fossil fuel burning or volcanic eruptions, chemically reacts to form sulfuric acid aerosols. Other sulfur compounds such as COS and CS_2 also react to form aerosols. Because of potential fuel strategy changes it is important to understand the present distribution of aerosols and sulfur gases in the atmosphere. With our capability to measure gas and aerosols in the stratosphere, it was natural for EML to develop the capability of measuring sulfur bearing compounds. We are presently the only research group in the world providing hemispheric distributions of COS in the stratosphere. The importance of these measurements to sulfur modeling is supported by the recent request for these data by researchers in the U. S. and Germany. Sulfate measurements, carried out by Los Alamos and previously the National Center for Atmospheric Research provide a ten year record of continuous sulfate measurement in the stratosphere.

Carbon Compounds in the Stratosphere

Carbon monoxide, CO_2 , and CH_4 are three additional compounds measured in the program. Both CO and CH_4 are important in the photochemistry of the stratosphere. It has been shown that the concentration of CH_4 is increasing in the atmosphere and could also affect the radiation budget of the earth. Carbon dioxide is another compound which is increasing at an alarming rate and could affect the earth's climate. Our capability allows us to make measurements of these gases in the upper troposphere and stratosphere and evaluate temporal changes in these compounds.

Volcanic Eruptions

Over the last few years HASP has been operational at the time when volcanic eruptions injected significant amounts of material into the atmosphere (Mt. St. Helens and El Chichon). Aerosols in the stratosphere can potentially absorb or scatter electromagnetic radiation from the sun or earth, possibly leading to climatic changes. The effect of natural and anthropogenic emissions on the atmospheric particle loading must be fully understood, in order to estimate the impact on climate. Volcanic eruptions emit enormous quantities of gases and aerosols into the atmosphere. We need to understand how these emissions affect the global aerosol burden. HASP has the capability of assessing the impact that volcanic eruptions and anthropogenic emissions have on atmospheric loading in the upper troposphere and stratosphere.

In 1980 we were able to sample in the plume from the eruption of Mt. St. Helens. Because of our capability to measure both radioactivity and stable constituents in the plume, we showed that the eruption did not inject megacuries of radon into the stratosphere as was reported by Fruchter et al. (1980) in Science. In addition we were able to characterize the aerosol composition in the plume and provide data on the sulfuric acid and ash content (Leifer, et al., 1981).

When El Chichon erupted we were in the middle of a regularly scheduled Airstream cycle and were able to intercept the plume from the eruption as the ash and gases circumnavigated the earth. From our measurements we were able to confirm that negligible amounts of COS were emitted by the volcano. The sulfate data provided a direct estimate of amount of sulfate injected into the stratosphere.

Arctic Haze

We have recently completed a mission with NOAA to evaluate the composition of Arctic haze. It has been shown that the haze is dominated by anthropogenic emissions containing sulfate, graphitic carbon and organic compounds. These compounds directly affect the radiation budget in the Arctic and could change the regional temperature. Characterization of both gases and aerosols are needed to understand the location of the sources. With our capability to fly at a range of altitudes, we are able to assess the vertical extent of the haze and provide background measurements in the upper troposphere and stratosphere for comparison.

Future Objectives

The future non-nuclear objectives for HASP lie in three areas:

- (1) continued measurements of aerosols and gases in the troposphere and stratosphere as a function of latitude and season,
- (2) improve our sampling capability in remote regions where concentrations of gases and aerosol are extremely low,
- (3) develop the capability to measure SO_2 and CS_2 gases and perform bulk chemical analysis in impactor samples and,
- (4) increase the number of researchers involved in sampling on the aircraft and balloon.

The unique and proven capability that the HASP provides for systematic sampling and analysis of pollutants in the stratosphere, dictates that this program should be continued indefinitely.

References

Fruchter, J. S., et al., Mount St. Helens Ash from the May 18, 1980 Eruption: Chemical, Physical, Mineralogical, and Biological Properties, Science 209, 1116-1125 (1980)

Leifer, R., L. Hinchliffe, I. Fisenne, H. Franklin, E. Knutson, M. Olden, W. Sedlacek, E. Mroz and T. Cahill, Measurements of the Stratospheric Plume from the Mount St. Helens Eruption: Radioactivity and Chemical Composition, Science 214, 904-907 (1981)

Molina, M. J. and F. S. Rowland, Stratospheric Sink for Chlorofluoromethanes: Chlorine Atom Catalyzed Destruction of Ozone, Nature 249, 810-812 (1974)

ATMOSPHERIC DEPOSITION

Herbert W. Feely
Donald C. Bogen

For at least 30 years EML has been engaged in the study of atmospheric deposition, mainly as part of its investigation of the fate of radionuclides injected into the atmosphere by nuclear weapons tests. Additionally during the past seven years EML has invested a significant effort in the measurement of the chemical composition of wet and dry deposition. This shift in emphasis was brought about in response to the growing awareness in the scientific community that the increasing use of traditional sources of energy in this country may be causing changes in the chemical composition of precipitation which could be damaging to important sectors of the environment, such as lacustrine biota, agricultural soils and forests.

It has long been known that the combustion of fossil fuels injects acidic gaseous pollutants, particularly sulfur dioxide and nitrogen dioxide, into the atmosphere (Smith, 1852; 1872). Coal combustion especially has been recognized as a major source of sulfur emissions (Smith, 1872; Crowther and Ruston, 1911). Ultimately these gaseous pollutants react with materials of the surface of the earth, becoming a component of the dry deposition, or are incorporated by chemical and physical reactions into liquid or solid constituents of the atmospheric aerosol. Aerosol particles are soon returned to the surface of the earth by wet deposition in rain or snow, or by dry deposition by impaction or sedimentation on vegetation or the soil.

Beginning in the 1950's, investigations in Europe linked the acidification of lakes in Scandinavia to the deposition of wind-borne acidic pollutants transported northward from industrial regions of Europe (Oden, 1968). Since the 1960's similar studies in the northeastern U. S. and eastern Canada have concluded that lakes in those regions have undergone acidification, with resultant damage to aquatic life (Hultberg and Stenson, 1970; Likens et al., 1972). Acidic rainfall has also been implicated in a number of other observed or hypothesized environmental problems. These include the corrosion of building materials (Oden, 1968), the decreased growth rate or death of trees, especially at

high altitudes (Jonsson, 1972), the possible destruction of soil microorganisms with resultant disruption of the geochemical cycles on which the growth of vegetation depends (Tyler, 1972; 1976), and possible deleterious effects on human health through the introduction of heavy metals into the food chain (Huckabee and Blaylock, 1974; Tyler, 1980). Several studies have related the chemical composition of precipitation to the trajectories followed by the air which yielded the precipitation. These studies have commonly concluded that acidic precipitation in the northeastern U. S. usually falls from air masses which have passed over the mid-western U. S. in the vicinity of the large coal-burning power plants of the Ohio River Valley. As a result there has been increasing political pressure to force industrial emitters of sulfur dioxide, and especially the older coal-burning power plants to reduce their sulfur emissions (Hileman, 1983).

The measures proposed to reduce industrial emissions, and thus presumably to reduce the incidence and severity of acidic rainfall, could involve costly investments in aging power plants. Consequently it is desirable that as much information as possible on the sources of the acidity in acid rain in the eastern U. S. be obtained before the federal government decides which steps must be taken to ameliorate the problem. The Interagency Task Force on Acid Precipitation was created by the Acid Precipitation Act of 1980 (P.L. 96-294) to coordinate federally funded research related to the problem of acidic precipitation. The EML studies of the chemical composition of deposition have been designated by DOE as part of that agency's involvement in the National Acid Precipitation Assessment Program, which was organized by this task force.

At the time that the EML study of the chemical composition of deposition began in mid-1976, no nation-wide networks for deposition sampling were as yet operational. It was planned that EML would establish such a network, so that data could be obtained which would characterize the variations in the chemical composition of both wet and dry deposition in a number of geographic regions which would differ from each other in climate and in the amount of acidic emissions which were being produced by the burning of coal, of oil and of other fuels. It was believed that the data so obtained could serve to test hypotheses concerning the effect that changing patterns of fuel use would have on the chemical composition

of deposition. Such data would also indicate the geographic extent of the regions receiving acidic deposition. Sampling was begun during 1976 and 1977 at 9 sites, including clean air sites, such as Mauna Loa Observatory, and polluted sites, such as New York City. Expansion of the network ceased when it became evident that the National Atmospheric Deposition Program (NADP), with its nationwide sampling network, would in fact be established. The EML network, as such, was abandoned in mid-1981, for by then the NADP network was fully operational and covered all of the regions covered by our network. Upon the basis of the information which had been obtained during the five years of existence of the EML network, new goals were set for the EML research effort. One of these goals is the characterization of the chemical composition of precipitation at several sites in the Metropolitan New York area which appear to experience different impacts from local sources of pollution. A second goal is the delineation of the relationship between the physical and chemical composition of the atmospheric aerosol and the composition of the dryfall collected in the dry side of a wet/dry collector.

Our current studies of the composition of precipitation were inspired by the observation that during 1977 and 1978 the concentration of sulfuric acid in precipitation at our sampling site on Manhattan Island in New York City was more than 50% higher than the concentration at the Chester, NJ site, only 58 km to the west. If all of the sulfuric acid in the precipitation at Chester and New York City were attributable to the long range transport of sulfate or sulfuric acid from distant sources, such as the mid-western U. S., we would expect the concentrations of this constituent in precipitation at those two sites to be approximately the same. When the concentrations at the two sites differ significantly as they did in 1977 and 1978, we must assume that some local source of sulfur pollution has affected disproportionately the New York City site. Most of the potential damage to structural materials by the action of acidic precipitation will occur in urban areas such as New York City. We believe, therefore, that a more detailed investigation of the possible impact of local sources upon the acidity of precipitation in the New York City region is justified. To provide more detailed information on the spatial distribution of acidity in precipitation falling in the metropolitan region, two additional sites were established, one in northern New Jersey, essentially between Chester and the City, and one within the City but about 15 km east of the Manhattan site.

A major goal of the EML research on dry deposition is the evaluation of the potential usefulness of the wet/dry collector for monitoring rates of dry deposition. It is known that both gaseous and particulate atmospheric pollutants will undergo dry deposition to the surface of the earth. Models of the transport and deposition of sulfur dioxide and sulfate and of nitrates and nitric acid indicate that dry deposition may account for half or more than half of the total deposition of these pollutants. Yet there is no technique available which has been shown to be capable of monitoring rates of dry deposition accurately and routinely at a network of stations. Without such a technique it is not possible to accumulate the data needed by those who are modeling the atmospheric transport and deposition of acidic pollutants to evaluate adequately the rates of dry deposition over large regions of the country.

The conventional wisdom states that the wet/dry collector cannot yield useful information about dry deposition because it is completely unlike the natural surfaces of soil and vegetation. But two lines of evidence support the potential usefulness of such collectors. The first is derived from past calculations of the deposition rate of radioactive fallout from nuclear weapons tests. A technique for making such calculations involved the measurement of the stratospheric inventories of radioactive debris at the beginning and end of a particular time interval, and calculating the fallout deposition from the decrease in the inventory. Another technique involved the integration of measurements of deposition samples obtained during this same time interval using collectors of a type similar to the wet/dry collector. The results of these two techniques generally agreed closely. A second line of evidence is derived from our participation in two phases of the Dry Deposition Intercomparison Study at Champaign, IL in September 1981 and in June 1982. We measured daily samples of dryfall obtained using six wet/dry collectors. Our estimations of sulfate deposition velocities during the 1982 experiment, at least, fall within the same range as do those reported by Wesely and coworkers using an eddy correlation technique (Wesely et al., 1983). The eddy correlation technique is based on physical principles and should give fairly accurate estimates of deposition velocities, but it is time-consuming and can only be used under optimum conditions of terrain and weather, so it cannot be used practically as a monitoring technique.

Our current study of dry deposition seeks to characterize the chemical and physical properties of the aerosol at a particular site (our site at Chester, NJ), and the properties of the dryfall collected by the wet/dry collector at the same site, and to determine whether there is a predictable relationship between them. We expect soon to reestablish the sampling of sulfur dioxide at this site also. Such sampling will enable us to determine whether this gas undergoes dry deposition into the wet/dry collector buckets, perhaps by reaction with previously deposited alkaline particles. We also plan to measure a number of similar dryfall samples to be collected at West Point, NY during the testing of a NOAA/EPA/USGS size selective sampler of aerosols and gases. This sampler is being evaluated under the direction of Dr. Bruce Hicks of ORNL for possible use at sampling sites of the National Trends Network to provide information from which rates of dry deposition will be calculated.

If rates of dry deposition can be calculated from samples obtained using the wet/dry collector, the calculation should be more straightforward, and perhaps will involve fewer assumptions, than if these rates must be calculated from aerosol data.

References

- Crowther, C. and H. G. Ruston, The Nature, Distribution and Effects upon Vegetation of Atmospheric Impurities in and near an Industrial Town, *J. Agr. Sci.* 4, 25-55 (1911)
- Hileman, B., Acid Rain: A Rapidly Shifting Scene, *Environ. Sci. Technol.* 17, 401-405 (1983)
- Huckabee, J. W. and B. G. Blaylock, Microcosm Studies on the Transfer of Hg, Cd, and Se from Terrestrial to Aquatic Ecosystems, D. D. Hemphill, ed., Proceedings Univ. of Missouri 8th Annual Conf. on Trace Substances in Environmental Health, Columbia, MO, pp. 219-222 (1974)
- Hultberg, H. and J. Stenson, Effects of Acidification on the Fish Fauna of Two Small Lakes in Bohuslan, South-western Sweden, *Flora Fauna* 1, p 11 (1970)
- Jonsson, B. and R. Sundberg, Has the Acidification by Atmospheric Pollution Caused a Growth Reduction in Swedish Forests", Research Note 20, Dept. of Forest Yield Research, Royal College of Forestry, Stockholm, Sweden (1972)
- Likens, G. E., F.H. Bormann and N. M. Johnson, Acid Rain, *Environment* 14, 33-40 (1972)

- Oden, S., The Acidification of Air and of Precipitation and its Consequences in the Natural Environment, Ecology Committee Bulletin No. 1, The State National Science Research Council, Stockholm, Sweden (1968)
- Smith, R. A., On the Air and Rain of Manchester, Mem. Lit. Phil. Soc., Manchester, Series 2, 207-217 (1852)
- Smith, R. A., Air and Rain: The Beginnings of Chemical Climatology, Longmans, Green, London, (1872)
- Tyler, G., Heavy Metals Pollute Nature, May Reduce Productivity, Ambio 1, 52-59 (1972)
- Tyler, G., Heavy Metal Pollution, Phosphatase Activity, and Mineralization of Organic Phosphorus in Forest Soils, Soil Biol. Biochem. 8, 327-332 (1976)
- Tyler, G., Metals in Sporophores of Basidiomycetes, Trans. Brit. Myc. Soc. 24, 41-49 (1980)
- Wesely, M. L., D. R. Cook and R. L. Hart, Eddy-Correlation Measurements of the Dry Deposition of Particulate Sulfur and Ozone at the Intercomparison Experiment, (abstract), Trans. American Geophysical Union 64, 198 (1983)

ENERGY-RELATED POLLUTANTS IN ECOSYSTEMS

Merrill Heit

Introduction

Several classes of pollutants have been found to be present in fossil fuels that are toxic or potentially toxic to humans and/or biota (Morris et al., 1980). Included are: 1) trace elements such as As, Be, Cd, Cu, F, Hg, Ni, Pb, Sb, Se, Sn, Tl, and Zn (Heit, 1977); 2) natural radionuclides including those in the ^{238}U and ^{232}Th series (Beck et al., 1980); and 3) organic compounds formed during incomplete combustion of fossil fuels, including those having mutagenic or carcinogenic properties such as the polycyclic aromatic hydrocarbons (PAH) and related heterocyclic compounds containing nitrogen and sulfur (Braunstein et al., 1977).

Since an expansion of coal-related generating stations and coal-related technologies has been predicted to meet the energy requirements of the United States in the near and long term future, increased amounts of these toxic and potentially toxic trace substances are expected to be released into the environment (Morris et al., 1980; NAS, 1981). We believe that it is relevant and useful to develop and carry out investigations which address the environmental questions associated with energy-related, biologically significant substances, released into the environment as a consequence of the combustion and/or production of fossil fuels, most notably coal. The information derived from this investigation should be applicable toward assessing the routes leading to human exposure as well as the amounts which reach people. Almost all of the contaminants investigated are cited by the U. S. EPA as being priority pollutants (Callahan et al., 1979). The substances are also stable in the environment and are generally associated with the respirable fraction of the aerosol produced during incomplete combustion of fossil fuels (Natusch, 1978). Most are said to show at least some "bio-accumulation" potential (Braunstein et al., 1977). It is our intent to measure and provide data on energy-related pollutants as they exist in the environment. We maintain that this approach is significantly more appropriate than "laboratory simulations" where synergistic and/or antagonist mechanisms, as they occur in the real world, cannot be realistically approximated.

Initial emphasis has been placed on measuring particulate associated energy-related pollutants rather than gaseous species since the former are more apt to be stable and hence provide a longer lasting record in the environment. Particulates are also more likely to be incorporated into food-chains.

According to the most recent recommendations of the U. S. Interagency Committee on Health and Environmental Effects of Energy Technologies (FIC, 1980), composed of representatives from DOE, HHS, and EPA, as well as recommendations by the former U. S. ERDA, Division of Biomedical Research (DBER, 1977) two of the most significant areas of environmental research related to the useage and production of fossil fuels are: 1) Chemical Deposition, including so called "acidic deposition" with its associated environmental stresses and 2) Effects on Human Health, caused by particulate-associated contaminants in respirable size. Research performed at EML in the program described herein directly addresses some of the most important questions posed within these two broad areas. Specific questions addressed are grouped into 4 main areas:

A. Deposition History and Assessment of Future Trends - What are the past, present and expected inputs of toxic and potentially toxic energy-related trace substances deposited into various regions of the U. S.? Are the inputs and concentrations of these substances significantly different between regions? What is the significance of local versus long-distance transport of these substances? How are these trends related to historic and projected patterns of energy useage? How do anthropogenic inputs of energy-related substances compare with deposits from natural or "biogenic" sources?

B. Human Exposure and Accumulation in Food-Chains - What are the energy-related contaminants which are of the utmost concern in regard to potential environmental transport and incorporation in food chains? Are any of these trace substances being incorporated into food-chains that can lead to humans? At the concentrations present in the environment do these substances pose potential health or ecological hazards? How long may we expect these pollutants to remain active in aquatic and terrestrial ecosystems, and are they changed in the environment into other species? What are the chemical and "biotransformation" dynamics of energy-related substances in sediments?

C. "Acidic" Deposition - Are the levels present in "acidic ecosystems" different from those found in neutral systems and are energy-related pollutants more likely to be incorporated into food-chains in these altered ecosystems? What are the potential health effects of energy-related pollutants deposited in acidic aquatic ecosystems? When did "acidification" of aquatic ecosystems begin and did observed changes occur under natural or anthropogenic influences? Are there consistent spatial and temporal trends of anthropogenic pollutants deposited in acidified lakes located in various regions of the U. S. ?

D. Methods Development - Are there biological monitoring protocols or indicator organisms that can be developed that will permit accurate assessment of the potential effects of releases from fossil fuel related technologies? In addition, are there indicator inputs or elements that may be used for exposure assessment and/or predictors of health effects including cancer? Are methods available for measuring biologically significant, energy-related pollutants with high accuracy and precision? Are current methods of estimating pollutant deposition histories correct - if not what is/are the preferred method(s) for estimating recent geo-chronologies?

Research aimed at obtaining answers to these questions has been stated to be of importance by several government and academic research organizations. Specific examples of agencies and organizations that have directly expressed interest in all or portions of this research include but are not limited to the following: a) The U. S. Interagency Task Force On Acid Precipitation composed of representatives of eleven federal agencies including DOE-ER. Major portions of this research have been given the highest priority by this organization and it is an intrinsic part of the National Acid Precipitation Assessment Plan (NAPAP) (NAPP, 1983; Altshuller, 1983); b) The National Atmospheric Deposition Plan (NADP) (Galloway et al., 1980); c) Tennessee Valley Authority (TVA, 1982); d) DOI-National Park Service (Heit, et al., 1983); e) EPA (Powers, 1983); f) NOAA (Heit et al., 1983); g) The New York State Department of Environmental Conservation (Heit and Klusek, 1979; Heit et al., 1981; Heit and Klusek, 1983); h) The N.J. State Departments of Health and Meadowlands Development Commission (EML, 1983a); i) The Electric Power Research Institute (EPRI) (Heit et al., 1983; Galloway et al., 1983; EML, 1983b); j) Several academic institutions

including the Woods Hole Oceanographic Institution (EML, 1983c), the University of Virginia (Galloway et al., 1982), and Cornell University (EML, 1983d).

Program Description

EML's program has been designed to provide meaningful information on many of the questions stated in the introduction. This is currently accomplished by centering the program about three main themes: Sediment History, Food Chains, and Methods Development.

Sediment History - Lake and reservoir sediments offer a unique substrate for investigating the occurrence of many energy-related pollutants deposited in the environment since they are the main "sink" for materials entering watersheds and may be dated by radioactive methods so as to provide a chronological history (Heit and Burke, 1977). In order to properly assess geo-chronologies of sediments in a meaningful way, methodologies must be available for taking large volume, undisturbed sediment cores so that multiple parameter analysis can be performed at critical core depths. We have such devices in our recently developed large diameter, slow penetration sediment corer (Burke et al., 1980) and 22' portable research vessel, HASL II (Burke et al., 1983). This equipment allows us to take large, undisturbed lake sediment cores and measure increments of sediment as thin as 1 cm for a suite of trace substances and environmental tracers indicative of natural and anthropogenic origins, as well as materials which are toxic to biota and/or humans.

Emphasis is placed on evaluating pollutant histories in ecosystems believed to receive their inputs via long distance atmospheric transport. Comparisons are made with sites receiving inputs directly from significant fossil-energy requiring sources located in the vicinity of the ecosystems under examination. Sites investigated for long-distance atmospheric inputs include 13 acidic and near neutral lakes and reservoirs in the N. Y. Adirondack Preserve and N. H. White Mountains; 5 reservoirs in the Southern Appalachian Mountains, some of which exhibit morphological anomalies in their resident fish populations that are believed to be caused by inputs of energy-related trace substances (Tennessee Valley Authority, 1983); 5 high altitude remote lakes in the Rocky Mountains of Colorado and 6 lakes and reservoirs in the Wasatch and Uintah Mountains of Utah

that are believed either to be susceptible to "acidic deposition" or else thought to receive significant atmospheric inputs from fossil-fuel requiring facilities. Results to date for these and related studies are shown in Heit, et al. (1983); Heit, et al. (1983); Galloway et al. (1983); Heit and Tan (1979); Heit (1979); Heit, et al. (1980); Tan and Heit (1981a) and Tan and Heit (1981b).

Measurements of pollutant histories at sites receiving inputs from local sources, here coal-fired power plants, are centered about studies at Cayuga Lake in the New York Finger Lakes Region in the eastern U. S. and Lake Powell on the Arizona-Utah border in the western U. S. Results to date for these and related studies are presented in references (Heit and Klusek, 1979; Heit and Tan, 1979; Heit et al., 1980; Klusek et al. 1983; Heit, in press 1983). Emissions from coal-fired power plants are significant in that most if not all of the pollutants released are also associated with other fossil-fuel technologies, although their relative abundances will vary. Thus, the study of coal-related pollutants may serve as a working model for the other fuel technologies.

Food-Chains - Representative biota from different trophic levels known or expected to act as biological accumulators of pollutants are collected and analyzed. For the present, emphasis is placed on aquatic systems, thus integrating with our sediment geo-chronological studies in that contaminants showing significant trends in the sediments are measured in the biota. Contaminants are also measured in the water from these systems and where appropriate "bio-accumulation potentials" are ascertained. Since a major goal of our research is to assess exposure of energy-related pollutants to humans through food-chains, emphasis is placed on measuring concentrations in edible tissues, i.e., muscle. Less importance is placed on the measurement of these pollutants in organs not usually eaten by humans, such as fish liver, even though they are documented accumulators of trace elements and organic compounds. Results to date for these studies are shown in references (Heit, 1979; Heit and Klusek, 1983). In research related to these studies, efforts have been underway aimed at evaluating selected organisms, most notably freshwater mussels, to serve as "indicator organisms" for assessing transformation and accumulation of energy-related substances deposited in aquatic ecosystems. Results to date are presented in references (Heit and Klusek, 1979).

Methods Development - A significant effort is placed in evaluating new methods for analyzing environmental samples with high accuracy and precision. To date, the majority of this research has centered on overcoming matrix interferences and evaluating appropriate analytical techniques for measuring trace elements in sediments and biota (Heit, 1977; Klusek and Heit, 1979; Klusek and Heit, 1981; Heit and Klusek, 1982; Klusek and Heit, 1982). Work has also been instituted aimed at developing methods for measurement of nitrogen and sulfur substituted PAH and other biologically significant compounds in environmental samples for which there are no reference materials. Since our laboratory is only equipped with a packed-column GCMS which has greatly reduced sensitivity and ability to discriminate between compounds, efforts are underway toward obtaining a capillary-column GCMS which will allow detection and discrimination of these environmentally significant molecules that are now impossible with the present apparatus. Methods have been developed at EML for measuring unsubstituted PAH in sediments (Tan, 1979).

Efforts are also underway at EML aimed at evaluating current procedures used by the scientific community to date sediment layers in cores for establishing geo-chronologies. This is of great significance since the results may help to firmly establish such important matters as the advent of "acidic precipitation" in North America.

Future Studies

As of September 1983, we have completed 75% of our goal of obtaining representative sediment cores from regions of the mainland U. S. deemed to be susceptible to ecological effects resulting from chemical deposition including "acid precipitation". It is expected that sediment cores will be taken from the Cascade and Sierra Nevada Mountains within the next few years. It has been suggested that these areas have received enhanced deposits of atmospherically transported, energy-related contaminants in recent times, which may have resulted in deleterious effects to remote watersheds (California Air Resources Board, 1981; Lewis and Grant, 1980). Until that time, we intend to continue to answer the questions set forth in the introduction using the comprehensive collection of sediment and biological samples now available for the northeastern, southeastern, and central and southwestern United States.

Work will continue aimed at establishing whether biologically active organic compounds associated with energy processes, are significant in food chains leading to man. As new questions arise regarding energy-related substances in the biosphere, additional analyses will be performed on archival samples and when appropriate additional samples will be obtained.

References

- Altshuller, A. P., The Acidic Deposition Phenomenon and its Effects: Critical Assessment Review Papers, Vol. II, Effects Sciences Public Review Draft, USEPA Report EPA-600/8-80-016B (1983)
- Beck, H., C. V. Gogolak, K. M. Miller and W. M. Lowder, Perturbations on the National Environment due to the Utilization of Coal as an Energy Source, in Natural Radiation Environment III, T. F. Gesell and W. M. Lowder, editors, pp. 1521-1558, CONF-780422, USDOE Symposium Series (1980)
- Braunstein, H. M., E. D. Copenhaver and H. A. Pfuderer, Environmental, Health and Control Aspects of Coal Conversion and Overview, ORNL Report No. ORNL FIS-95, Vol. 2 (1977)
- Burke, J. C., W. R. Clarke, M. Heit and H. L. Volchok, A 22 Foot Portable Research Catamaran: Background Design and Use to Date, USDOE Report EML-381, 17-41 (1980)
- Burke, J. C., R. E. Hamblin and S. A. Casso, Tripod Modification of Sphincter Corer: Construction Operation, Core Extrusion and Sampling Efficiency, Woods Hole Oceanographic Institution Report (in press, 1983)
- California Air Resources Board, Proceedings of Californis Symposium on Acid Precipitation, San Francisco, CA, Jan. 14-15, 1981, pp 1-279 (1981)
- Callahan, M., et al., Water-Related Environmental Fate of 129 Priority Pollutants, Vol. 1 and Vol. 2, USEPA Report EPA-440/4-79-029, Washington, DC (1979)
- Division of Biomedical and Environmental Research, R.I. Van Hook and W. D. Shults, Editors, in Proceedings of a Workshop, U.S. Energy Research and Development Administration Report ERDA 77-64, pp. 1-79 (1977)
- EML (1983a) EML has taken sediment cores from Berry's Creek, Rutherford, NJ in order to evaluate the possible association of atmospherically deposited energy-related trace substances in a statistically significant leukemia "pocket" thought to be affecting a public school district in N.J. (1983a)

EML (1983b) EML is also associated with the on-going Regional Integrated Watershed Assessment Study (RILWAS) now taking place in 10 Acidic Adirondack Lakes. Sediment cores and water samples are being evaluated. Several Universities, including Indiana, Cornell, Colgate, Smith, Syracuse, Maine, and RPI are involved in this EPRI coordinated study (1983b)

EML (1983c) EML is part of intercalibration studies for measuring energy-related organic pollutants (PAH) in biota. This research is coordinated through Woods Hole Oceanographic Institution. EML has also provided Woods Hole scientists data on sediment cores used for assessing recent historical inputs of radioactivity on several occasions (1983c)

EML (1983d) Because of our expertise in sediment coring, EML has been asked by Cr. Gene Likens to take sediment cores from Mirror Lake, NH to firmly establish the geo-chronologies and anthropogenic histories in the Hubbard Brook region of N. H.

Federal Interagency Committee on the Health and Environmental Effects of Energy Technologies, Health Effects of Coal Technologies Research Needs, The Mitre Corp., Reports No. MTR-79W 15902, pp. 1-201, McLean, VA (1980)

Galloway, J. N., S.J. Eisenreich and B. C. Scott, Toxic Substances in Atmospheric Deposition: A Review and Assessment, National Atmospheric Deposition Program Report NC-141, Jekyll Island, GA (1980)

Galloway, J. N., D. Banning, A. D. Prete, M. Heit and A. O. Davis, A Summary of Observations for the Investigation of the Sediments of Panther, Sagamore and Woods Lakes, in Electric Power Research Institute Report, R. E. Goldstein, Ed. The Integrated Lake-watershed Acidification Study: Proceedings of the ILWAS Annual Review Conference, EPRI Report EA-2827 (1983)

Galloway, J. N., J. D. Thornton, S. A. Norton, H. L. Volchok and R. A. McLean, Trace Metals in Atmospheric Deposition, A Review and Assessment Atmospheric Environment 16, 1677-1700 (1982)

Heit, M., A Review of Current Information on Some Ecological and Health Related Aspects of the Release of Trace Metals into the Environment Associated with the Combustion of Coal, USERDA Report HASL-320, pp I-51 (1977)

Heit, M., C. S. Klusek and J. Baron, Evidence of Deposition of Anthropogenic Pollutants in Remote Rocky Mountain Lakes, Water, Air and Soil Pollution (in press, 1983)

Heit, M., Y. L. Tan and L. E. Toonkel, An Evaluation of Methodologies for Measuring Polycyclic Aromatic Hydrocarbons (PAH) in Sediment, USDOE Report EML-390, 213-235 (1981)

- Heit, M., Variability of the Concentrations of Seventeen Trace Elements in the Muscle and Liver of a Single Striped Bass, *Morone Saxatilis*, Bull. Environ. Contam. Toxicol, 23, 1-5 (1979)
- Heit, M. and C. S. Klusek, The Release of Trace Substances by a Coal-Fired Power Station into an Aquatic Ecosystem 1-Environmental Sampling in Cayuga Lake, NY, USDOE Report EML-363, pp 3-30 (1979)
- Heit, M., Y. Tan, C. Klusek and J. C. Burke, Anthropogenic Trace Elements and Polycyclic Aromatic Hydrocarbon Levels in Sediment Cores from Two Lakes in the Adirondack Acid Lake Region, Water, Air, Soil Pollution 15, 441-464 (1981)
- Heit, M. and C. S. Klusek, Trace Elements Concentrations in the Dorsal Muscle of White Suckers, *Catostomus Commersoni* and Brown Bullheads, *Ictalurus Notatus* from Acidic Adirondack Lakes, Water, Air and Soil Pollution (in press, 1983)
- Heit, M., Y. Tan, C. Klusek and J. C. Burke, Trace Elements and Polycyclic Aromatic Hydrocarbon Levels in Sediment Cores from Sagamore and Woods Lakes. Electric Power Res. Inst. Report, R. E. Goldstein, Ed. The Integrated Lake - Watershed Acidification Study: Proceedings of the ILWAS Annual Review Conf. EPRI Report EA-2827, Palo Alto, CA (1983)
- Heit, M. and J. C. Burke, Sediment Sampling in Six Western Lakes, USERDA Report HASL-315, 83-91 (1977)
- Heit, M. and Y. L. Tan, The Concentration of Selected Polynuclear Aromatic Hydrocarbons in the Surface Sediments of Some Fresh and Marine Waters of the United States, USDOE Report EML-353, 3-39 (1979)
- Heit, M., The Concentrations of Benzo(a)pyrene in the Sediments of Six Western United States Lakes, Water, Air, and Soil Pollution 15, 441-464 (1979)
- Heit, M., Y. L. Tan, C. S. Klusek, H. L. Volchok and J. C. Burke, The Origin and Deposition History of Trace Elements and Polynuclear Aromatic Hydrocarbons in Two Remote Lakes in the Adirondack Acid Rain Region, USDOE Report EML-381, pp. I-75 - I-127 (1980)
- Heit, M., C. S. Klusek and H. L. Volchok, Time History of Trace Elements in Sediments from Standley Lake, CO, Environmental International 4, 229-239 (1980)
- Heit, M., The Relationship of a Coal-fired Power Plant to the Levels of Polycyclic Aromatic Hydrocarbons in Cayuga Lake, Water, Air, and Soil Pollution (in press, 1983)
- Heit, M., C. S. Klusek and K. M. Miller, Trace Elements Radionuclide and Polynuclear Aromatic Hydrocarbon Concentrations in Unionidae Mussels from Lake George, Environmental Science and Tech. 14, 465-468 (1980)
- Heit, M., An Interlaboratory Comparison of Trace Elements Analyses of a Near Shore Marine Sediments, USDOE Report HASL-328, 25-37 (1977)

- Heit, M. and C. Klusek, The Effect of Dissecting Tools on the Trace Elements Concentrations of Fish and Mussel Tissues, *Science of the Total Environment* 24, 129-134 (1982)
- Klusek, C. S. and M. Heit, Effect of Different Preservation Methods on the Trace Elements Concentrations of Fish and Mussel Tissues, *Bulletin of Environmental Contamination Toxicology* 28, 202-207 (1982)
- Klusek, C. S., K. M. Miller and M. Heit, Trace Elements and Radionuclide Mass Balances at a Coal-fired Electric Generation Station, *Environ. International* 9, 139-144 (1983)
- Klusek, C. S. and M. Heit, An Intercomparison of the Analyses of Marine Sediments for Fifteen Trace Elements, USDOE Report EML-363, 31-85 (1979)
- Klusek, C. S. and M. Heit, An Intercomparison for the Analyses of Commercially Available Reference Materials for 22 Trace Elements, USDOE Report EML-390, 91-211 (1981)
- Lewis, W. M. and M. C. Grant, Acid Precipitation in the Western United States, *Science* 207, 176-177 (1980)
- Morris, S. C., et al., Health and Environmental Effects of The National Energy Plan - A Critical Review of Some Selected Issues, Biomedical and Environmental Assessment Division, National Center for Analysis of Energy Systems, Brookhaven National Lab., Report BNL 51300, Upton, NY (1980)
- National Academy of Sciences, Atmosphere-Biosphere Interactions: Toward a Better Understanding of the Ecological Consequences of Fossil Fuel Combustion, National Academy Press, Washington, DC pp. 1-263 (1981)
- National Acid Precipitation Program, Effects Research Review, USEPA/NCSU Acid Precipitation Program, North Carolina State University - M. Heit, Task Group Project E1-10, Time Histories of Toxic Substances in Acidified Aquatic Ecosystems and M. Heit, Task Group Project E4-3, Toxic Substances in the Biota of Acidified Aquatic Ecosystems. Presented at North Carolina State University Acid Deposition Program, Feb. 21-25, 1983 (1983)
- Natusch, D. F. S., Potentially Carcinogenic Species Emitted to the Atmosphere by Fossil-Fuel Power Plants, *Env. Health Perspect.* 22, 79-90 (1978)
- Powers, C. E., Personal Communication, USEPA Environmental Research Laboratory, Corvallis, OR. EML is providing information on the water chemistry of several remote Western lake believed to be susceptible to acidic precipitation) (1983)

Tan, Y. L. and M. Heit, Biogenic and Abiogenic Polynuclear Aromatic Hydrocarbons in Sediment from Two Remote Adirondack Lakes, *Geochimica Cosmochimica Acta* 45, 2267-2279 (1981a)

Tan, Y. L. and M. Heit, Analysis of Polynuclear Aromatic Hydrocarbons in Sediment Cores from Two Lakes in the Adirondack Acid Rain Belt by High Resolution Gas Chromatography/Mass Spectrometry, in M. Cooke and A. J. Dennis (Editors), *Chemical Analysis and Biological Fate: Polynuclear Aromatic Hydrocarbons - Fifth International Symposium*, Battelle Press, Columbus, Ohio (1981b)

Tan, Y. L., Rapid Simple Sample Preparation Technique for Analyzing Polynuclear Aromatic Hydrocarbons in Sediments by Gas Chromatography-Mass Spectrometry, *J. of Chromatography* 176, 319-327 (1979)

Tennessee Valley Authority, Southern Appalachian Acid Precipitation Research Plan - Aquatic Assessment for the Blue Ridge Province, U. S. TVA, Chattanooga, TN. (EML will provide the deposition history into the Southern Appalachian Mountains from power production in the Tennessee Valley including Northern Georgia. Determinations will be made of the causes of morphological anomalies in fish species found in selected reservoirs.) (1982)

QUALITY ASSURANCE PROGRAMS AT EML - OVERVIEW

Herbert L. Volchok

EML's quality assurance (QA) activities grew out of a clearly recognized need, for better knowledge of the quality of the measurements and data produced in environmental, ecological and health studies. DOE strategies in production, distribution and conservation of energy could have deleterious effects on health through changes in pollutant levels in the biosphere. The accuracy and precision with which potential health effects can be predicted is directly dependent on the accuracy and precision of the measurements of the pollutants in question, in any and all compartments of the environment and the biosphere. How good are the measurements? In a broad sense, that is the question QA must answer.

For more than two decades the Laboratory has gradually and increasingly become involved in a variety of activities involving quality control/quality assessment/quality assurance in a wide variety of environmental applications relevant to the DOE-OHER-EML interests (e.g., Harley et al., 1975; Harley 1980; Sanderson et al., 1980; Volchok et al., 1980). Several of the write-ups and presentations in this Program Review touch upon or make reference to some of these activities (e.g., see submissions of Beck, Hardy et al., Krey, George). In addition three of the Laboratory's QA programs which have broad national/international implications, the Quality Assessment Program and the radon and TLD intercomparisons, were chosen for more detailed coverage (see submissions by Sanderson, de Planque, Fisenne). All of the Laboratory's QA efforts are described in the EML Annual Report for 1982 (Volchok et al., 1983). In the following I will summarize a number of these activities.

One grouping of these activities concerns participation on committees or working groups engaged in the preparation or writing of standard procedures and methodology in both nuclear and non-nuclear physics and chemistry. At present a number of EML staff are actively engaged in programs with organizations such as the American Society for Testing Materials (ASTM), the National Commission for Radiation Protection (NCRP),

the American Nuclear Society (ANS), the American National Standards Institute (ANSI), the International Organization for Standardization (ISO) and the International Electrotechnical Commission (IEC).

Another category of QA effort at the Laboratory involves preparation of analytical standards and reference materials for development of analytical methods and for quality control purposes. Most recently, in response to perceived needs in the analytical community and a request from colleagues, we developed a ^{99}Tc vegetation reference material (Feiner, 1983). About 400 samples of this material are currently available upon request. A program that has continued over several years concerns preparation of "natural matrix" standards (NMS). This grew out of recommendations of the International Committee for Radiation Metrology (ICRM). Working closely with the National Bureau of Standards (NBS) we have completed Standard Reference Materials (SRM) for several types and activity levels of soil and sediment, human liver and human lung (Volchok et al., 1980a and b; Volchok and Feiner, 1984). In addition, the Laboratory maintains an extensive library of nuclear and non-nuclear standards and reference materials (see submission of Volchok and de Planque) for internal use in procedures development, quality control and service to the QAP (see submission of Sanderson). In this program we also make available standards and tracers to U. S. and foreign laboratories with related research activities.

The Laboratory has always participated in selected national and international laboratory intercomparisons as validation of internal QA and in certification procedures for standards development. Typically this involves between five and ten such intercomparison exercises per year. Occasionally, we have been asked to carry out an intercomparison effort among DOE research contractors as an indication of the quality of the analyses being reported and interpreted (Volchok and Feiner, 1979).

The Offsite Radiation Exposure Review Project (ORERP) is a study of the radiation exposure of the population of some western states from fallout originating in the nuclear weapons tests at the Nevada Test Site in the 1950's (see submission of Beck). The contractors carrying out the program, which includes: site selection, in situ gamma spectrometry, soil sampling, sample preparation and analyses for ^{137}Cs and plutonium, are

using systems and techniques developed by EML over the many years of our studies in the region. For this reason EML was asked to develop a QA protocol for the ORERP and assist in its application.

Summarized above are the highlights of EML's major QA programs, exclusive of the three separately presented. There appears to be increasing awareness of the value and necessity for sound and thorough QA in all laboratories and similar institutions and we cannot therefore predict other than that the EML activities in this field will continue with probable demand for expansion.

References

- Feiner, M. S., A ⁹⁹Tc Vegetation Reference Material (in press, 1983)
- Harley, J. H., H. L. Volchok and V. T. Bowen, Quality Control in Radiochemical Analysis in: Reference Methods for Marine Radioactivity Studies II, IAEA Tech. Report Series, No. 169, pp. 63-68, Vienna (1975)
- Harley, J. H., Experience with an Analytical Quality Control Program, Efficient and Environmental Radiation Surveillance, ASTM STP 698, pp. 257-269, American Society for Testing and Materials (1980)
- Sanderson, C. G., L. K. Cohen, A. Goldin, A. N. Jarvis, L. Kanipe, C. Sill, M. Trautman and B. Kahn, Quality Assurance for Environmental Monitoring Programs, in: Upgrading Environmental Radiation Data Health Physics Society Committee Report HPSR-1, p. 5-1 (1980)
- Volchok, H. L., G. de Planque and C. Sanderson, QA Programs at the Environmental Measurements Laboratory, Proc. of Second International Conf. on Low Radioactivities 1980, p. 85-96, 1980, VEDA Publishing House of the Slovak Academy of Sciences (1980a)
- Volchok, H. L. and EML Staff, Environmental Measurements Laboratory 1982 Annual Report, USDOE Report EML-410 (1983)
- Volchok, H. L., M. S. Feiner, K.G.W. Inn and J.F. McInroy, Development of Some Natural Matrix Standards, Progress Report, Env. International 3, 395-398 (1980b)
- Volchok, H. L. and M. S. Feiner, Natural Matrix Radioactivity Standards and Reference Materials, Nuclear Instruments and Methods (in press, 1984)
- Volchok, H. L. and M. S. Feiner, A Radioanalytical Laboratory Intercomparison Exercise, USDOE Report EML-366 (1979)

THE EML-OOS QUALITY ASSESSMENT PROGRAM

Colin G. Sanderson

U. S. Department of Energy contractors handling hazardous materials are required to maintain environmental monitoring programs which assess the actual or potential danger of exposure to these materials. In order to make these assessments, environmental samples such as water, soil, vegetation, tissue and air particulates collected on filter papers must be obtained, prepared, and eventually submitted for analysis. However, inherent limitations of analytical methods produce data which are subject to variations and uncertainties. Since the resulting data become the basis for determining health risks, a program to assess their accuracy is very important.

DOE's Office of Operational Safety (OOS), recognizing the need for an external assessment of the quality of environmental analyses, assigned the responsibility for developing a program to EML because of its unique position in the analytical community. The Laboratory:

- (1) has a proven expertise in the field of analytical radiochemistry,
- (2) has the expertise to prepare appropriate environmental reference materials in the large quantities required for the QAP,
- (3) has adequate experienced staff to perform the multiple measurements required, and
- (4) has a recognized record of impartiality.

The semi-annual reports published by EML, provide Headquarters and DOE Field Offices with the data they require to determine the validity of environmental reports issued by their contractors.

The QAP was initiated in 1976 and ran smoothly for five years. Because of organizational and budget constraints within DOE, the program was terminated in 1981. However, it was soon recognized by OOS that the need for quality assessment of the contractors data was not being met. It was also recognized that EML was the only laboratory that could satisfy all the requirements of the program at reasonable cost. The program was re-instituted at EML in November 1982.

At the present time EML collects, processes and analyzes the radiochemical concentrations in intercomparison samples which are currently distributed to about thirty participating laboratories twice a year. These samples include most matrices encountered in environmental monitoring, namely air filters, aqueous solutions, soil, vegetation and tissue. A report comparing the participating laboratories' results to EML's values is issued for each sample distribution (Sanderson, 1983). In addition a statistical evaluation of the reported data is published annually (Sanderson, 1981). An annual workshop is held for participating laboratory, DOE Field Office, DOE Headquarters, and EML personnel to evaluate the program and make necessary adjustments if required.

In FY-84 we will begin preparing samples for non-nuclear analyses which will be distributed in FY-85. Future expansion of the program will be based upon the needs of the participants and OOS.

While we do not believe participation in this program should be a Laboratory's only quality assurance effort, it satisfies a minimum requirement for a blind, external and impartial assessment necessary for legal and scientific credibility.

References

- Sanderson, C. G., Semi-Annual Report of the Department of Energy, Operational Safety, Health and Environment Division - Quality Assessment Program, USDOE Report EML-417 (1983)
- Sanderson, C. G., Summary Report of the Department of Energy, Division of Operational and Environmental Safety Quality Assurance Programs 13 and 14, USDOE Report EML-388 (1981)

RADON INTERCOMPARISONS

Isabel M. Fisenne

The limited potential energy available from fossil fuel sources and the relative scarcity of such sources prompted a number of nations in Western Europe to pursue conservation measures as national policy. The oil embargo by OPEC nations directed U. S. policy toward conservation of energy. Tax credits were offered to industry and dwelling owners who retrofitted equipment and physical structures to reduce energy losses. This "tightening" of structures was based on the fact that the heat loss from a unit was reduced by decreasing the number of air turnovers per hour within a unit.

With the decrease in the turnover rates within structures, it was found that pollutant concentrations in the indoor atmosphere increased. As early as 1956, Hultquist showed decreasing ventilation rates increased the indoor concentration of the airborne natural radionuclides, ^{222}Rn , ^{220}Rn and their daughter products (Hultquist, 1956). Although it has long been recognized that ^{222}Rn gas contributes a negligible fraction to the alpha radiation dose to the human lung (Harley, 1953), the presence of an elevated radon concentration at a particular site warrants measurements of radon daughters.

Major radon surveys of housing have been performed in Canada and Sweden (McGregor et al., 1980; Hildingson, 1982) while the principle U. S. studies of radon and daughter concentrations in homes have focused on relatively small geographical areas of naturally or technologically enhanced radioactivity. Examples of such programs are those in the Florida phosphate area and the western uranium mining areas.

Essentially no national data base exists which documents the indoor radon and daughter concentrations from which risk estimates for human health effects can be derived for the general population. Without empirical information of known analytical quality, assessment of the impact of conservation measures on the life-time risk for the induction of lung cancer from the inhalation of radon and its daughters would be based on the few studies of limited scope currently available.

Radon daughter exposures of 100 working level months (WLM) have been shown to produce an excess number of lung cancers in uranium miners (Radford, 1981). The life-time risk for the induction of lung cancers in this occupationally exposed group is about 2×10^{-4} per WLM. NCRP estimates that the exposure of the U. S. population averages 0.2 WLM per year or about 10 to 20 WLM life-time exposure. However, certain situations could expose members of the general population to cumulative life-time exposures in excess of 100 WLM. Corrective action may be warranted in such situations but no action could be initiated without verifying the situations from reliable empirical information.

Several U. S. federal agencies support, either directly or through grants to national laboratories, universities and private sector laboratories, programs aimed at the measurement of radon and its daughters in occupational, remedial action and environmental settings. In all these cases, radon (not daughters) is measured. The validity of pooling the data collected by a number of investigators performing measurements in different locations is dependent upon some knowledge of the quality of the measurement data. Evaluations based on poor quality information may lead to economically wasteful decisions and perhaps result in unnecessary deleterious human health effects.

EML has a 35 year history in quality measurements of radon at occupational and environmental concentrations. EML possesses the unique combination of a primary radon measurement facility and an controlled environmental radon calibration chamber. These elements have been used in tandem for research projects, instrument and dosimeter calibration and evaluations. Peer recognition of this special combination has essentially led to the establishment of the EML facilities as a de facto calibration facility for radon studies in the United States and in several European nations.

EML organized an intercomparison program for evaluating the quality of radon measurements. One of the primary purposes of these exercises is the development of empirical evidence for determining the validity of pooling data collected by various groups in an effort to form a uniform data base for dosimetric considerations. In addition these intercomparison exercises are cost-effective by coordinating quality control efforts among laboratories through a single agent.

Twenty different facilities actively engaged in the field collection and measurement of radon have participated in five EML intercomparison since April 1981. Although not involved in field measurements the National Bureau of Standards participated in two intercalibrations. In all, 6 U. S. Federal government laboratories, 2 National Laboratories, 3 private sector laboratories, 2 State laboratories, 4 Universities, 2 European institutes and a Canadian laboratory have joined in these exercises. Summary results of the exercises have been published in two EML reports (Fisenne et al., 1981; Fisenne et al., 1983) and a peer review journal note (Fisenne et al., 1983).

We expect the EML radon intercomparison exercises to continue in response to the needs of the scientific community to maintain and improve the quality of radon measurements for at least as long as energy conservation efforts continue to change the indoor environment.

References

- Fisenne, Isabel M., Andreas C. George and Mark McGahan, Radon Measurement Intercomparisons at EML, USDOE Report EML-397 (1981)
- Fisenne, Isabel M., Andreas C. George and Helen W. Keller, The 1982 Radon Inter-comparison Exercises at EML, USDOE Report EML-413 (1983)
- Fisenne, Isabel M., Andreas George and Mark McGahan, Radon Measurement Inter-comparisons, Health Phys. 45, 553-556 (1983)
- Harley, John H., Sampling and Measurement of Airborne Daughter Products of Radon, Nucleonics 11, 12-15 (1953)
- Hildingson, O., Radon Measurements in 12,000 Swedish Homes, Environment International 8, 67-71 (1982)
- Hultqvist, Bengt, Studies of Naturally Occurring Ionizing Radiations with Special Reference to Radiation Doses in Swedish Houses of Various Types, Kungl. Svenska Vetenskapsakademiens Handlingar, 6 (1956)
- McGregor, R. G., P. Vasudev, E. G. Le Tourneau, R. S. McCullough, F. A. Prantl and H. Taniguchi, Background Concentrations of Radon and Radon Daughters in Canadian Homes, Health Phys. 39, 285-289 (1980)
- Radford, E. P., Radon Daughters in the Induction of Lung Cancer in Underground Miners, Banbury Conference Report No. 9 (1981)

TLD INTERCOMPARISONS

Gail de Planque

With the advent of the atomic age came a continuously growing interest in, and concern for, radiation doses received by the general public from both natural and anthropogenic sources. Environmental dosimetry evolved as a discipline especially in response to these concerns. Advances in the state-of-the-art of environmental dosimetry proceeded rapidly in the 1970's as the expansion of the nuclear power industry accelerated the need for environmental monitoring programs around these facilities as well as the need for studies of the natural radiation environment to aid in the interpretation of the measurements. Today, the most prevalent detectors used for environmental dosimetry are solid state thermoluminescence dosimeters (TLD's).

By the early 1980's, the dosimetry community had witnessed:

1. one or more TLD monitoring networks around every operating nuclear power plant and most nuclear facilities in the United States (Conti, 1978) and around many, if not most similar facilities in the rest of the world;
2. the recent establishment by the U. S. Nuclear Regulatory Commissions of its own TLD monitoring network at all nuclear power plants nationwide (Slobodien and Cohen, 1980; Mattson, 1980);
3. routine use of TLD's for studying environmental radiation, a recent example being the study of a high background area in the People's Republic of China and consequent health effects (High Background Radiation Research Group, China, 1980);
4. progress in both national and international standards for environmental thermoluminescence dosimetry (de Planque and Gesell, 1982);
5. reports of a variety of environmental applications at the series of International Conferences on Luminescence (more recently, Solid State) Dosimetry (see for example the Proceedings of the Fifth International Conference on Luminescence Dosimetry, 1977, and the Proceedings of the Sixth International Conference on Solid State Dosimetry, 1980; and,
6. an assessment of the population dose and health impact of the accident at the Three Mile Island Nuclear Power Plant based largely on environmental TLD measurements (Battist et al., 1979).

EML has been heavily involved since the 1960's in programs directed at the development of environmental thermoluminescence dosimetry. These efforts have included the design and construction of a TLD reader (Volchok and de Planque, 1982), numerous studies of the natural radiation environment using TLD (Gulbin and de Planque, 1983; de Planque, 1980), and studies of both measurement techniques (Burke, 1976) and data interpretation methods (de Planque, 1979) for nuclear facilities monitoring.

It was this experience and the results of a pilot study involving five laboratories (Dickson et al., 1975), that generated a discussion of the overall problems of environmental dosimetry at a workshop held at EML, then the U. S. Atomic Energy Commission Health and Safety Laboratory (Lowder, 1974), and eventually led to the formation of an ad hoc committee for the purpose of carrying out more comprehensive and systematic studies of environmental dosimetry. The result was the initiation of a series of full-scale international intercomparisons of integrating dosimeters, primarily TLD's, each organized in cooperation with one or two collaborators.

The series began in 1974 and there have been six intercomparisons to date with the seventh being planned for summer, 1984 (Gesell et al., 1976; de Planque and Gesell, 1979; Gesell et al., 1978; Gesell and de Planque 1980; de Planque and Gesell, 1982; Gesell et al., 1983). The intercomparisons draw wide international participation with one hundred and thirty-four laboratories in thirty-one countries from North and South America, Europe, Africa and Asia sending dosimeters for the most recent study. They typically include about fifteen different types of commercial or homemade dosimeters, and readers from almost all of the commercial manufacturers in the world as well as a wide variety of non-commercial systems.

The intercomparisons essentially evaluate the performance of integrating environmental radiation dosimeters under both field and laboratory conditions, in a manner appropriate to assess their performance for applications such as environmental monitoring around nuclear facilities and studies of the natural radiation environment. They enable an assessment of the state-of-the-art mainly by the dispersion of the results, the comparison of the results with estimated delivered doses, and the correlation of the results with variables such as the type of dosimeter system, method of calibration, and general practices and procedures. To ensure on-going consistency in these performance

evaluations, the protocol has remained basically the same, however, each study has included a unique procedure or test designed to investigate a particular problem associated with environmental dosimetry. Hence in addition to being an international quality assessment program, the intercomparisons incorporate a research effort on a scale essentially impossible to achieve in any other way available in practice due to the extensive numbers and types of TLD systems involved.

The intercomparison results are usually analyzed to determine the performance of the participating laboratories in terms of the criteria in American National Standard on environmental applications of TLD (ANSI, 1975 (1982)), and participants frequently report their results as evidence of the quality of the performance of their own systems.

We expect to continue these studies as long as there remains a need for evaluating the state-of-the-art and for providing a mechanism for individual laboratories to assess the quality of their environmental measurements. Following the trend established in the area of personnel dosimetry (ANSI, 1982), a new U. S. standards work group, N13.29 has begun work on developing a standard on "Criteria for Testing Environmental Dosimetry Performance." The standard will specify tests, test conditions, and performance criteria for dosimetry systems, mainly TLD, that are used to measure integrated exposure or absorbed dose of γ -radiation at levels as low as natural background under out-of-doors environmental conditions normally encountered in the United States. It will essentially be based on the intercomparison studies and is expected to delineate a continuing testing and certification program for environmental dosimeters which regulatory agencies may use as a basis for judging the acceptability of dosimetry programs used for environmental measurements, especially around nuclear facilities. If such a program is eventually established, it may be incorporated with the present intercomparison studies, or be used instead at least for U. S. dosimetry services. Such a program would not, by definition, provide a complete replacement for the intercomparisons due to its more limited scope and number and variety of participants.

References

- ANSI N545-1975(82), American National Standard Performance, Testing, and Procedural Specifications for Thermoluminescence Dosimetry (Environmental Applications), American National Standards Institute, New York (1975 and 1982)
- ANSI N13.11, Criteria for Testing Personnel Dosimetry Performance, American National Standards Institute, New York (1982)
- Battist, L., J. Buchanan, F. Congel, C. Nelson, M. Nelson, H. Peterson and M. Rosenstein (ad hoc Population Dose Assessment Group), Population Dose and Health Impact of the Accident at the Three Mile Island Nuclear Station, U. S. Government Printing Office, Washington, DC (1979)
- Burke, G. de P., Thermoluminescence Dosimetry: Environmental Monitoring Near Nuclear Reactor Sites, IEEE Trans. on Nuclear Science, NS-23, 1224-1236 (1976)
- Conti, E. F., Radiological Environmental Monitoring by NRC Licensees for Routine Operations of Nuclear Facilities, U. S. Nuclear Regulatory Commission Staff Task Group, USNRC Report NUREG-0475 (1978)
- de Planque, G., Evaluation of Methods for the Determination of X- and Gamma-Ray Exposure Attributable to a Nuclear Facility Using Environmental TLD Measurements, USNRC Report NUREG/CR-0711 (EML-355) (1979)
- de Planque, G., TLD Measurements and Model Calculations of Environmental Radiation Exposure Rates, in: Proc., The Natural Radiation Environment III, T. F. Gesell and W. M. Lowder, Eds., USDOE Report CONF-780422, Vol. 2, pp. 987-1003 (1980)
- de Planque, G. and T. F. Gesell, Second International Intercomparison of Environmental Dosimeters, Health Physics 36, 221-233 (1979)
- de Planque, G. and T. F. Gesell, Thermoluminescence Dosimetry - Environmental Applications, Int. J. Appl. Radiat. Isot., 33, 1015-1034 (1982)
- Dickson, H. W., F. F. Haywood and K. Becker, Tenth Dosimetry Intercomparison, Oak Ridge National Laboratory Report ORNL-TM-4566 (1975)
- Fifth International Conference on Luminescence Dosimetry, Sao Paulo, Brazil, Physikalisches Institut Justus-Liebig-Universitat Giessen, Heinrich-Buff-Ring 16, D 6300 Giessen (1977)
- Gesell, T. F. and G. de Planque, Highlights of the Fourth International Intercomparison of Environmental Dosimeters under Field and Laboratory Conditions, Nuclear Instruments and Methods 175, 186-188 (1980)

- Gesell, T. F., G. de Planque Burke and K. Becker, An International Intercomparison of Environmental Dosimeters, *Health Physics* 30, 125-133 (1976)
- Gesell, T. F., R. E. Gammage and G. de Planque, The Third International Intercomparison of Environmental Dosimeters under Field and Laboratory Conditions, *Health Physics* 35, 909 (1978)
- Gesell, T. F., F. L. Kalbeitzer and G. de Planque, Highlights of the Sixth International Intercomparison of Environmental Dosimeters, *Proc. Seventh Intl. Conf. on Solid State Dosimetry*, Ottawa, Canada, Radiation Protection Dosimetry (in press, 1983)
- Gulbin, J. and G. de Planque, Ten Years of Residential TLD Monitoring., *Proc. Seventh Intl. Conf. on Solid State Dosimetry*, Ottawa, Canada, Radiation Protection Dosimetry (in press, 1983)
- High Background Radiation Research Group, China Health Survey in High Background Radiation Areas in China, *Science* 209, 877-880 (1980)
- Lowder, W. M., Ed., *Proc., Second Workshop on the Natural Radiation Environment*, USAEC Report HASL-287 (1974)
- Mattson, R., J. Millhoan, B. Purple, J. O'Reilly, J. Scinto, M. Minners, R. Scroggins, and B. Russell, NRC Action Plan Developed as a Result of the TMI-2 Accident, USNRC Report NUREG-0660, Vol. 1 (1980)
- Sixth International Conference on Solid State Dosimetry, Toulouse, France, *Nuclear Instruments and Methods* 175 (1980)
- Slobodien, M. J. and L. K. Cohen, A Nationwide Program for Environmental Radiation Monitoring Around Nuclear Power Reactors, *Health Physics* 39, 1027-1028 (1980)
- Volchok, H. L. and G. de Planque, Eds., *EML Procedures Manual*, pp. C-04-01 - C-04-11 (1982, this manual is updated annually)

INSTRUMENTATION DEVELOPMENT AND SUPPORT PROGRAMS

Robert T. Graveson

The instrumentation development and support programs are intimately related to the objectives of the Laboratory's present and future scientific programs. In general, the needs for a specific measurement capability evolves from the close collaboration between the scientist and the engineer. As new facets of a problem are revealed by scientific experiments, the objectives of instrument development change to encompass the new directions that may be necessary. The end result, often is not a specific instrument, but an effective measurement methodology tailored to the application. The engineering solutions to the problems of the experimenter may be based on the implementation of some commercially available equipment, with or without modification to adapt it to the application. If such is not available, a new instrument is designed and built in the prototypes facilities of the Laboratory.

The Instrumentation development and support efforts provides several services within the scope of the Environmental Measurements Laboratory's scientific programs. The staff involved:

- a) Develop new techniques for the detection of environmental pollutants.
- b) Provide expert advice to the scientific staff on questions of measurement techniques, as well as electronic and mechanical methodology, and computer based data systems.
- c) Provide design, fabrication, and repair services to maintain a supply of operational instrumentation for use in the scientific programs of the Laboratory.
- d) Design, equip, and operate experimental facilities for the diagnostic analysis and calibration of experimental and commercial instruments.

The following are some specific examples of the instrumentation development and support programs.

Radon Instrumentation

Radon and the associated daughter products are significant contributors to the radiation dose in man. The short half lives of the daughters while resulting in high specific

activity, require that concentration measurements be made in the area of exposure. Thus, a portion of the effort has been devoted to developing instrumentation for the scientific projects which is easily transported, yet still highly reliable. The Time-Integrating Environmental Radon Daughter Monitor (Guggenheim et al., 1979) is based on the use of thermoluminescent dosimeters as the integrating detector. This unit has been used for both residential and "tailing pile" studies for a number of years. A new version (EML Type TF-11) (Latner, et al., 1981) uses a semiconductor detector, and the integration to determine average exposure is provided by electronic digitized memory. This unit is approximately ten times more sensitive than the earlier TLD design. An accessory printing scaler permits time synoptic data to be collected for the analysis of the changes in exposure levels in these applications. In collaboration with the scientific staff a battery-operated portable alpha counter and a portable scaler conforming to their requirements for light weight, yet delivering laboratory quality performance, were designed and have found a place in the field studies of the Laboratory.

Several years ago, the Laboratory needed a reliable monitor for establishing environmental levels of radon. The resulting automatic monitor was installed at the baseline station in Chester, NJ so that the natural variations at that site could be characterized over an extended period. This system is based on the filter collection of the radon daughters resulting from radon decay in a one hundred liter chamber. After a collection period of sixty minutes, the filter is transported to a counting position under a scintillation counter. This unit has exceptional statistical accuracy and a successful operational history over several years.

Electrostatic fields have been used as a means to collect radon daughters for the detection of radon. While this approach produced a convenient, light-weight unit, there was a suspicion that the daughter collection might depend on the relative humidity. Since this effect could influence the accuracy of estimating radon exposure, a study was started to determine the electrical characteristics of radon daughters. Calibrations under closely controlled conditions of exposure to a moist atmosphere were used to document the influence of humidity (Watnick et al., 1976). Since collection efficiency depends on the electrical charge of the radon daughter molecules, several experiments were designed

to elucidate this parameter. It was determined that in dry air most of the molecules were positively charged. The effect of moisture is to increase recombination, thus reducing the number of charged molecules (Negro and Watnick, 1978; Watnick and Negro, 1978).

Gamma Instrumentation

The dose delivered by environmental gamma-ray background has been the subject of many studies conducted by EML documenting the natural radiation environment. To differentiate small variations in the "normal" level, either from place to place or as a function of time and changing conditions, necessitates a unique degree of instrument stability. The MOSFET electrometer was developed for these types of instruments and is both time and temperature stable over an extended range. The best known, and most widely used, of these applications is the high pressure ionization chamber based Environmental Gamma Dose Rate Monitor. Commercial versions of this design are used extensively for off-site monitoring near nuclear power plants. A portable version, the SPICER (EML Type TN-8) (Latner et al., 1983), has been developed for use in the Laboratory's studies of environmental dose rates.

There has been an interest in using some type of low level instrument which has a range of energy independence that includes the x-ray region. The Kolb-Lautenbach scintillation probe achieves $\pm 15\%$ energy independence from 25 KeV to 1 MeV through the combination of a plastic phosphor and a zinc sulfide coating. An instrument, the KLERUS (EML Type TN-1), has been designed which uses this detector. The circuit incorporates the temperature independent electrometer configured as a current-to-frequency converter. The storage and display sections of this unit utilize conventional low-power integrated circuits.

Special Environmental Test Facilities

The Environmental Measurements Laboratory has a number of unique facilities for simulating controlled environments. These systems are utilized by many of the research projects and are essential for calibrating instruments and for developing new measurement techniques. In these facilities response parameters can be determined for stable

concentrations of several materials under known conditions of temperature, humidity, and physical abuse. These provide a means of evaluating new concepts in instrumentation development as they would perform in practical applications.

The High Altitude Test Chamber (HATCH) can reproduce the temperature and pressure conditions representing any altitude from sea level to over 150,000 feet. The large room inside the chamber comfortably accommodates any size sampling equipment which might be installed in high-altitude aircraft or suspended from a balloon. Thus, it has been, and continues to be, essential for determining the calibration of sampling flow rates, collection efficiencies, and system reliability of equipment used for stratospheric sampling. These parameters are difficult and often impossible to determine through "on-board" measurements, yet are necessary for use in the interpretation of the collected samples.

The Radon Calibration Chamber has an internal volume which approaches that of an average room. Air samples can be taken from this large volume without influencing the radon and radon daughter concentrations. The atmosphere within the chamber is maintained at radon levels from fractional picocuries per liter to concentrations up to one hundred pC/liter. Temperature and humidity also can be controlled over the usual environmental range. Currently, this unit is the basis for the Laboratory's International Intercalibration Program and is used for the calibration of EML's radon instrumentation.

An unusual facility is the Low Background Steel Room, where the gamma-ray background is significantly lower than exists in the natural environment. For instruments, and monitors, intended to establish the usual levels of exposure to the general population, calibration of their response down below the levels they can be expected to detect is an important part of their performance tests. Also, for some types of measurement system, the low levels of environmental gamma flux that can be achieved allow cosmic-ray response to be established. In addition, the chamber provides an unusual "test bed" for the development of new analytical concepts. Gamma-ray detector and spectrometer concepts can be studied, modified, and improved without the time and expense of providing the usual external "background" shielding structures.

The Regional Base Line Station at Chester, New Jersey, is located in an undisturbed area that is representative of Eastern Rural United States. Sampling and in-situ monitoring

systems are establishing the normal levels of both radioactive and non-nuclear components of the atmosphere. This station, which is within seventy miles of EML, provides a contrast to the Metropolitan area as monitored at this Laboratory, which is located in the heart of New York City. The Chester station contains meteorological instrumentation, 80th Meridian Heavy-duty Air Samplers, a continuous Radon Monitor, and computer based data storage as its standard complement. Special experiments are conveniently installed, and a telephone data link allows frequent access to the progress of selected measurements.

Analytical Facilities

Over the years, several counting room facilities have been installed for the analysis of environmental samples. This equipment is routinely maintained and, as appropriate, it is updated with newer electronics and improved detectors.

The High Resolution Gamma Spectroscopy unit contains four germanium detectors, configured to meet the specialized needs of low activity environmental samples.

The Low Level Beta Counting Room is equipped with twenty cosmic-ray guarded scintillation counters and an automatic data acquisition system. These counters are based on an original EML design and combine high efficiency with a low counting background averaging 0.25 counts per minute, making these systems still a uniquely sensitive detector for strontium-yttrium 90.

The pulse ionization chambers are the basis for many of the radon standardizations. While other methodology now is used for the field measurements, these units provide low level, reproducible analyses. Ten chambers have been modernized in recent years and continue to supply the basis for most intercomparison calibrations.

Low Level Alpha Spectroscopy is the basis for plutonium counting. The silicon semiconductor detectors, with their combined highly specific energy resolution and their low inherent counting background, are satisfactory for the analysis of environmental samples.

Data Acquisition Systems

Many of the Laboratory's analytical measurements are involved with environmental samples, i.e., with samples containing very low concentrations. Thus, long counting

times are required to obtain adequate statistical precision. The continuous recording of data in long-term monitoring presents a similar situation where a large amount of information is acquired at low data acquisition rates. Automatic acquisition and storage of this information has two advantages. First, it minimizes the involvement of an operator when the data is being collected and, secondly, it allows the collected information to be analyzed by computer systems without necessitating the transcription of the numbers.

Microprocessor based electronics have made possible data acquisition systems which are flexible, reliable and yet effect considerable savings in both the space required and in the cost. Two of these module based designs which have been used extensively by the Laboratory are the SMART-1 (EML Type TL-12) (Polito and Negro, 1979) and the CLEVER Counter (EML Type TL-13) (Negro and Reiser, 1977; Negro, 1982). Usually, these units provide their collected digital data, in computer compatible form, on teletype punched paper tape, or on magnetic tape cassettes.

More recently, inexpensive, yet powerful, microcomputers have been used successfully for the acquisition, control and display of experimental data in a real-time, on-line mode. Our most common equipment is the Hewlett-Packard 85 small computer. Several of the routine applications are with the Thermoluminescent Dosimeter Readers, the Gas Chromatography Analyzers (Torquato, et al., 1982), the Atomic Absorption Spectrophotometer (Nagourney, et al., 1983), the Perfluorocarbon Analyzer of the field collected tracer transport experiments, and for the control of the Radon Calibration Chamber. Where appropriate, the microcomputers are connected to the Laboratory's main computer so that collected data can be transferred to the main data banks.

Thus, a portion of the Division's efforts are devoted to designing the electronic hardware and computer software necessary for interfacing analytical instruments to microcomputer systems.

High Altitude Sampling Program (HASP)

Two systems have been developed for use in WB-57 aircraft to sample stratospheric air. The Clean Air System (CLASS) (Leifer et al., 1981) consists of nine sampling flasks, the pressurizing pump, the inlet and outlet solenoid valve manifolds and the associated

control circuits. This unit is constructed of ultra-clean materials and to be contamination free.

Any materials which might absorb trace gases has been avoided. The result is a unit which has been shown to be effective for the collection of trace amounts of 15 to 50 compounds, such as the freons, CO₂, CO, methane, carbonyl sulfide, nitrous oxide, and others.

The particle collector system is a four stage inertial impactor design which draws samples through a probe protruding forward from the nose of the aircraft. The collector separates and collects particles as small as 0.05 micrometers diameter and determines number and size per unit volume. Unique collectors were developed to mount the oil surfaced grids on a drum type of sample changer. Analysis is for both chemical composition through Proton Induced X-ray Emission (PIXE) and by microscope. This impactor was useful in comparing quiescent conditions in the stratosphere with those subsequent to volcanic eruptions (Mt. St. Helens and El Chicon) when sulphuric acid was found dominant.

References

- Guggenheim, S. F., A. C. George, R. T. Graveson, A. J. Breslin, A Time-Integrating Environmental Radon Daughter Monitor, Health Phys. 33, 452-455 (1979)
- Latner, N., S. Watnick, and R. T. Graveson, Integrating Working Level Monitor EML Type TF-11, USDOE Report EML-389 (1981)
- Latner, N., K. Miller, S. Watnick and R. T. Graveson, Spicer - A Sensitive Radiation Survey Instrument, Health Phys. 44, 379-386 (1983)
- Leifer, R., K. G. Sommers, S. F. Guggenheim and I. Fisenne, Recent Developments in High Altitude Aircraft Sampling - Mt. St. Helens and Stratospheric Trace Gases, IEEE Transactions on Nuclear Science, NS-28 (1981)
- Nagourney, S. J., D. C. Bogen and V. C. Negro, Matrix Effects in the Determination of Mn and Pb in Atmospheric Deposition, Presented at Pittsburgh Conference and Exposition, March 1983
- Negro, V. C. and S. Watnick, Fungi - A Radon Measuring Instrument with Fast Response, IEEE Transaction on Nuclear Science, NS-25 (1978)

Negro, V. C. and T. Reiser, A Micro-Computer Data Logging System, in Micro-computer Design and Applications, S. C. Lee, Ed., pp. 163-193, Academic Press (1977)

Negro, V. C., Microcomputer Data Acquisition Module, in Microprocessor Application Handbook, Chapter 5, D. F. Stout, Ed., McGraw-Hill Book Co. (1982)

Polito, H. D. and V. C. Negro, SMART I and CLEVER Counter Environmental Data Acquisition Systems, IEEE Trans. on Nuclear Science, NS-26 (1979)

Torquato, C., N. Latner and D. C. Bogen, Further Developments in the Computer-ized Acquisition and Reduction of Ion Chromatographic Data, Proceedings of Rocky Mountain Conference, Denver, CO (1982)

Watnick, S., V. C. Negro and R. T. Graveson, Environmental Radon Monitor Varies with Relative Humidity, EML Technical Memo 76-14 (1976)

Watnick, S. and V. C. Negro, Radon Measuring Instruments - Electrostatic Field Collection, EML Technical Memo 78-3 (1978)

ANALYTICAL SUPPORT AND DEVELOPMENT

Philip W. Krey

Analytical support and development are necessary functions of an environmental research laboratory. These vital functions are important in both the planning and management of research programs.

In planning environmental research, multidisciplinary involvement is the most effective approach. This involvement at EML often embraces other disciplines as well as chemistry. Research plans formulated from such a broad scientific base are more extensive, thorough and commonly address areas of study closed to a single discipline.

Research goals often initiate the analytical research to develop chemical and instrumental methods needed to achieve those goals. However, once initiated, an iterative process develops in which improvements in the analytical technology permit research plans to be extended or redirected. This symbiosis is a healthy and constructive force in molding the research programs of the Laboratory.

A stable and experienced analytical research staff often anticipates the probable needs of future environmental studies and builds the technological foundations that will ultimately satisfy those needs. Fairly recent development of expertise in gas chromatography, ion chromatography, and atomic absorption spectrometry subsequently resulted in EML becoming a leading research laboratory in the fields of atmospheric tracing, atmospheric deposition and "acid rain" studies to name a few (Bogen et al., 1978, 1979, 1980; Ferber et al., 1981, 1982; Hefter et al., 1980; Nagourney and Bogen, 1978, 1981).

In management of environmental research, the need for on site analytical support and development is also readily recognized. Focused responsibility and rapid communication minimize administrative problems and maximize flexibility in pursuing unforeseen promising routes of study. Feedback from impartial analytical experts who are committed to the scientific investigation of a program and to the excellence of the Laboratory is a valuable resource in the management of a research program.

This recognized expertise and impartiality are also desirable characteristics of a laboratory called upon to assess the quality of the analytical work performed for the DOE by contractors. EML staff evaluates and recommends action to be taken on proposals to the Laboratory, OHER, or other components of DOE. EML developed and directs the Quality Assessment program which addresses the accuracy and precision of the analyses of environmental samples by about 30 DOE contractor laboratories (see submission by Sanderson).

The need for analytical support and development is more easily seen when the analytical requirements cannot be met in the private sector because of a lack of capability, stringent reporting schedules, or the national sensitivity of the work involved. For example, our analytical work in the atmospheric tracing program, rapid reporting of gamma spectrometric measurements in the High Altitude Sampling Program and the sensitivity of our ^{90}Sr analysis in diet and bone samples is not readily available in industry. However, the Laboratory has always had a policy to utilize commercial analytical services when they are available and meet requirements. In fact, EML has encouraged commercial analytical services through technology transfer by publishing and updating its Procedures Manual (The EML Procedures Manual, 1982) which has earned international acceptance.

This vital analytical support and development function can only be provided by a stable, impartial, well-equipped laboratory staff. The Analytical Chemistry Division occupies 12,000 square feet of laboratory space with both nuclear and non-nuclear capabilities. There are six standard laboratories, a class 100 air clean-room laboratory, a gas analysis laboratory, an instrumentation laboratory and four radioassay rooms. Involved in this effort are 20 full-time permanent personnel, including 16 professionals, 3 technicians, and 1 secretary.

Our analytical methods are state-of-the-art or on the leading edge of current research. To provide the needed support the Division is expert in gamma-ray and alpha particle spectrometry, low-level beta radioassay, radiochemistry in environmental samples, radon chamber measurements, gas chromatography, and gas-chromatography-mass spectrometry. The staff provides the analytical development and support function

to almost all of the energy related research programs of the Laboratory which are described in the other submitted material of this Laboratory review.

Our immediate goals are to continue in our present level of support. Our development interests are at once both broad and specific. We constantly review the scientific literature and the manufacturers' communications to keep abreast of all new analytical techniques and improved instrumentation. We are also working toward computer interfacing of all laboratory instrumentation to insure efficient and accurate data accumulation, reduction and reporting.

Specifically, we are planning to replace our older model GC-MS with an up-to-date capillary GC-MS to permit the analysis of the potentially carcinogenic nitrogen and sulfur containing polycyclic aromatic hydrocarbons in environmental samples. We are assembling two large diameter planar germanium detectors to view samples inserted between them which will improve the sensitivity of detection for low energy photons. For example, this configuration will markedly improve the quality of analysis of ^{210}Pb in lake sediment samples which can provide an estimate of the time when the various depths of the sediment were deposited. A large germanium well detector surrounded by a NaI anti-coincidence guard will be assembled and tested to detect coincident photon sum peaks, and thereby reduce the limits of detection for some short lived fission products. This system will upgrade our capability to detect and quantify the presence of nuclear weapons fallout from announced or clandestine tests. Both of these configurations represent advances in the field of gamma-ray spectroscopy which may provide the technology required to solve problems still on the horizon.

We plan to upgrade our X-Ray Fluorescence System to improve the sensitivity and efficiency in the analysis of atmospheric filter samples for selected trace metals. Work will continue in the use of analyte concentrating techniques to improve the sensitivity of atomic absorption spectrometry and ion chromatography. Further studies involving chemical modifiers or instrument adaptation will be directed toward eliminating sample matrix effects in electrothermal atomic absorption spectrometry.

Recent radiochemical studies at EML have shown that many of the historical analysis of soil samples collected near the Nevada Test Site (NTS) for $^{239+240}\text{Pu}$ and even ^{90}Sr have

underestimated their true values. This is believed to be due to the presence of a form of NTS fallout (conjectured to be a fused silicate particle) which does not yield its total radioactive content to the classical leaching methods. Procedures for the complete dissolution and analysis of 50 g samples of soil or larger for $^{239+240}\text{Pu}$ are almost complete. The resolution of this issue is of great importance to the success of the DOE's Offsite Radiation Exposure Review Project.

Because of its volatility and high mobility in soil and water systems, the long-lived fission product, ^{99}Tc has a relatively high potential for contaminating the environment around reprocessing plants and waste disposal sites. ^{242}Cm and ^{244}Cm are transuranics of high yield in high burnup nuclear fuel and are responsible for most of the alpha activity in the early years after extraction from the reactor. We consider it prudent to be prepared to investigate potential hazards to the population from these nuclides, as a result of operations or accidents, and therefore plans to develop procedures for their analyses in large environmental samples are underway. Intake and excretion patterns of natural environmental radionuclides in the general population are needed to estimate the impact of occupational exposure and will be useful also in assessing exposures to artificial sources. To address this issue we plan to develop a sequential scheme of isotopic analysis for uranium, thorium and radium. EML's sample library of earlier diet surveys in New York and San Francisco could then provide a wealth of high quality intake information for these natural alpha emitters.

References

- Bogen, D. C. and G. A. Welford, Radiometric Determination of Sulfate, *Journal of Radioanalytical Chemistry* 42, 307-312 (1978)
- Bogen, D. C. and S. J. Nagourney, Ion Chromatographic Analysis of Cations at Baseline Precipitation Stations, *Ion Chromatographic Analysis of Environmental Pollutants*, J. D. Mulik and E. Sawicki, Editors, Ann Arbor Science (1979)
- Bogen, D. C., S. J. Nagourney and C. Torquato, A Field Evaluation of the HASL Wet-Dry Deposition Collector, *Water, Air and Soil Pollution* 13, 453-458 (1980)
- Ferber, G. J., K. Telegadas, J. L. Hefter, C. R. Dickson, R. N. Dietz and P. W. Krey, Demonstration of a Long-Range Atmospheric Tracer System Using Perfluorocarbons, Final Report, Interagency Energy/Environment R & D Program Report, EPA-600/7-81-008 (1981)
- Ferber, G. J., K. Telegadas, C. R. Dickson, R. Lagomarsino, P. W. Krey and R. N. Dietz, Perfluorocarbon Tracer Experiments, ASCOT Data from the 1981 Cooling Tower Plume Experiments in the Geysers Geothermal Area, P. H. Gudiksen, Ed., Lawrence Livermore National Laboratory Report UCID-19332, ASCOT-82-4, Vol. II, 7-49 to 7-119 (1982)
- Hefter, J. L., G. J. Ferber and P. W. Krey, Atmospheric Tracer Experiments for Regional Dispersion Studies, *Proceedings of the European Seminar on Radioactive Releases and their Dispersion Following a Hypothetical Reactor Accident*, Riso, Denmark, Vol. II, pp. 953-969 (1980)
- Nagourney, S. J. and D. C. Bogen, Measurement of Trace Constituents in Global Baseline Precipitation Samples, *Canadian Journal of Spectroscopy*, 23, 101-104 (1978)
- Nagourney, S. J. and D. C. Bogen, Determination of Mn and Pb in Atmospheric Deposition at a Remote Coastal Site, *Water, Air, and Soil Pollution* 15, 425-432 (1981)
- The EML Procedures Manual, 25th Edition, Edited by H. L. Volchok and G. de Planque, Environmental Measurements Laboratory, U. S. Department of Energy, HASL-300 (1982)