EML-571

ENVIRONMENTAL MEASUREMENTS LABORATORY

1994 annual report

August 1995

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1994 annual report

EML

Environmental Measurements Laboratory

Philip W. Krey, Harold L. Beck, and Staff of the Laboratory

Nancy A. Chieco, Editor

U.S. Department of Energy
376 Hudson Street
New York, NY 10014-3621

August 1995
EML - 571

INTRODUCTION

This report summarizes the activities of the Environmental Measurements Laboratory (EML) for the calendar year 1994 and it serves as an annual report to the Director of the Office of Energy Research (ER), the Associate Director and staff of the Office of Health and Environmental Research (OHER), the manager and staff of the Chicago Field Office, and our colleagues. Emphasized are the progress and accomplishments of the year, rather than future plans or expectations. The technical summaries are grouped according to the following seven general program areas:

- Environmental Radiation and Radioactivity
- Radiation Transport and Dosimetry
- Environmental Radon, Thoron, and Related Aerosols
- Atmospheric and Surface Pollutant Studies Related to Global Climate Change
- Atmospheric Chemistry
- Metrology, Consultation, and Emergency Response
- Environmental Management

EML's mission is to address important scientific questions concerning human health and environmental impacts. Through its multidisciplinary staff, EML conducts experimental and theoretical research on radioactive and other energy-related pollutants, and provides DOE and other federal agencies with the in-house capability to respond effectively and efficiently with regard to quality assurance activities, environmental issues and related national security issues.

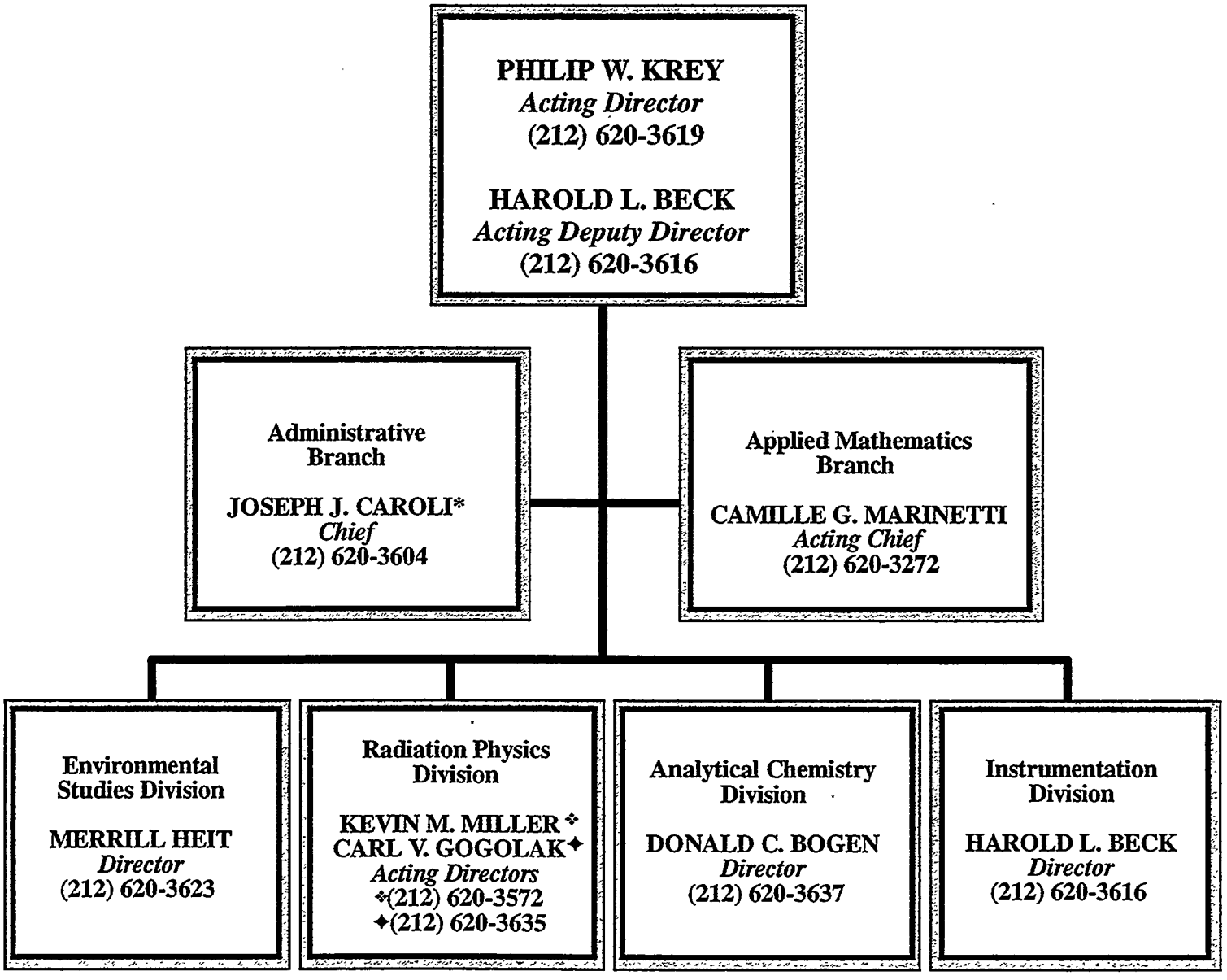
The Laboratory, now in its 47th year, is a government-owned, government-operated laboratory, programmatically under OHER in ER. The Laboratory is administered by the Chicago Field Office. At any given time there are typically 95-100 persons on board, including 89 FTEs (full-time equivalents), visiting scientists, and students working under the stay-in-school program. Of the FTE employees, 60 are scientific and 29 are professional and administrative support staff. The scientific staff consists of specialists in organic/inorganic/geo/radiochemistry; radiation/aerosol physics; biology; ecology; meteorology; mathematics; computer science; and mechanical/electrical engineering.

Further details about EML's history, organization, facilities, programs, and staff are contained in the EML brochure.

Listed at the end of the report are the Laboratory's seminar program, staff activities, publications, publications in progress, and presentations.

Inquiries and/or comments on any of the activities reported herein are invited.

ORGANIZATION AND STAFF



** Replaced Richard Tyson upon his retirement, September 30, 1994*

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* Replaced Richard Tyson upon his retirement, September 1994

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Departures

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Kostek, Alexander
Lowder, Wayne
Livingstone, John
Patterson, Tara
Romeo, Jeffrey
Stripp, Richard
Stuart, Robert
Tyson, Richard
Wilson, Frederick

CONTENTS

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Environmental Radiation and Radioactivity

1.1 OVERVIEW

Merrill Heit and Kevin M. Miller

The objective of the research performed in this program is to provide fundamental data on natural and anthropogenic radionuclides deposited on the earth's surface, which can be used to characterize, quantify and model environmental pathways, and to evaluate the environmental and human health impacts on regional and global scales. As part of this program, a rapid response capability is maintained to support Department of Energy (DOE) activities in radiation emergencies that arise within the U.S., and for assessing releases that are international in scope.

A significant component of the work performed includes the operation and maintenance of EML's global network (see Figure 1.1) comprised of over 115 sampling sites dispersed throughout the world. The EML network is continuously poised to instantly react to any new introduction of radioactivity into the environment anywhere on the globe due to the planned, clandestine or accidental detonation of a nuclear weapon a nuclear reactor accident (for example the Chernobyl tragedy), a transportation accident involving nuclear materials, or a space satellite accident during launch or atmospheric re-entry involving a nuclear reactor or a radioactive thermal generator.

Research related to the EML global network has lead to the development of state-of-the-art field radiation detection systems. These systems can measure air filter samples *in situ* for trace quantities of gamma-ray emitting radionuclides released into the atmosphere and transmit the data via satellite to a central location. Without these systems, ^{7}Be and other radionuclides with short half-lives decay in samples collected at remote or weathered-in stations before the samples can be returned to the laboratory for analyses. The deployment of these field radiation systems, called remote atmospheric measurements systems (RAMS), allow for the detection of these radionuclides at remote locations throughout the world. The resulting data are used to study the factors that affect the global distribution and temporal trends of natural and anthropogenic radioactivity in the lower troposphere.

Naturally occurring radioisotopes such as ^{7}Be and ^{210}Pb are also measured since they serve as natural tracers of lateral transport, vertical mixing, and wet scavenging processes, in the atmosphere. The atmospheric distribution of these substances therefore serves as a valuable test of the accuracy of meteorological models developed for global climate investigations. EML's global network database constitutes a major portion of the worldwide surface air database that is available for this research.

The use of EML's network data for atmospheric model verification and related climate issues is presented in the "Atmospheric and Surface Pollutant Studies Related to Global Climate Change" program area.

Another element of the Environmental Radiation and Radioactivity research program is to fully characterize background radiation fields in terms of spatial and temporal variations and to then assess increases due to human activities, particularly those that relate to DOE operations in such areas as energy development and site remediation. To this end, studies are performed that refine the state of knowledge on the source terms, transport, and accumulation of radioactive species in the environment.

Expertise is accrued in high precision measurements and instruments and techniques are developed with the aim of transferring this technology to DOE laboratories and their contractors, to other government agencies, or to the private sector. This includes developments in spectrometry, as well as total dose rate measuring instruments, such as pressurized ionization chambers and thermoluminescence dosimeters. Quality assessment projects that are international in scope are performed in the area of environmental radiation dosimeters.

In response to DOE's need for improved methods for site characterization in environmental restoration and waste management, applications of *in situ* techniques for determining radionuclide concentrations in soil and the adaptation of specialized detectors for this purpose are emphasized. The research reports in this section describe such activities that were performed in 1994. Other specific projects and applications are contained in Section 6 of this report.

A related area of research is to use rare stable and cosmogenically-produced isotopes as tracers of environmental processes. Thermal ionization mass spectrometry (TIMS) is used to measure individual isotopes of plutonium, uranium, neptunium and technetium to study their distribution and cycling within different components of ecosystems. Accelerator mass spectrometry (AMS) is used to measure cosmogenically and anthropogenically-produced radionuclides such as ^{36}Cl and ^{129}I that serve as time markers to estimate the rates at which complex environmental cycling occurs. The immediate goal of this research is to provide information on the behavior of subsurface waters at DOE production facilities and above-ground redistribution of radioactive contaminants at these sites. In each of these activities, basic information on the dynamics of the systems under study are required to adequately assess the environmental impacts of remediation strategies. Currently, investigations are conducted at the Idaho National Engineering Laboratory (INEL) on plutonium isotopes in soils.

The last component performed in this program is carried out to increase our knowledge of the pathways, magnitude, fluxes and inventories of pollutants that have been atmospherically deposited or released into diverse and/or ecologically complex systems, such as the Alaskan and Russian Arctic. Questions that need to be answered in this research include:

1. When did the pollutants begin to increase and at what rates?

2. How much has been deposited over the long term?
3. How do anthropogenic inputs of these substances compare with deposits from natural sources?

Determining the origin, fate and future trends of pollutant deposition are all components of this research. An additional portion of our Arctic research is part of a larger U.S. effort to determine if radioactive waste management practices of the former Soviet Union have potentially compromised fisheries - resources in the Arctic Ocean or, in any way, have led to radioactivity levels of concern in this ecosystem. The program is under the direction of the Office of Naval Research (ONR), and will continue through at least mid-1996.

Collaborative efforts with national and international organizations are performed on a continuing basis within this broad research area. EML is represented on committees for the International Commission on Radiation Units and Measurements (ICRU) and the American National Standards Institute (ANSI). We serve as consultants to the Nuclear Regulatory Commission (NRC) on a contractual basis whereby EML is developing the measurement and statistical testing methodology to support proposed decommissioning criteria that is based on a concept of "indistinguishable from background" (see Section 6 of this report for further information). EML has also been monitoring the Tokamak Fusion Test Reactor (TFTR) at the Princeton Plasma Physics Laboratory to provide fundamental information on the radiation environment around that facility during high yield DT reactions. The Institute of Electrical and Electronic Engineers, the Health Physics Society and the Harvard School of Public Health rely on EML scientists for the organization and teaching of short courses in radiation measurements. EML continues to be represented on special committees for the International Atomic Energy Agency (IAEA) (see Section 6 of this report), and we advise other government agencies and contractors on an informal basis in areas of mutual scientific interest.

Collaborative efforts are also carried out on the behavior of subsurface waters at DOE production facilities with scientists located at: the Department of Physics, Purdue University, IN; Nuclear Structure Research Laboratory, University of Rochester, NY; Los Alamos National Laboratories, NM; Argonne National Laboratory, IL; Pacific Northwest Laboratory, WA; U.S. Geological Survey, ID; and the IsoTrace Laboratory, University of Toronto, Canada. Scientific collaboration is also pursued with the Woods Hole Oceanographic Institution, University of Toronto, the U.S. Geological Survey, Oak Ridge National Laboratory, Argonne National Laboratory and Pacific Northwest Laboratory in the measurement of radionuclide releases from the former Soviet Union into Arctic seas. Research on pollutants deposited onto Alaskan and Russian Tundra and fresh water lakes has also been conducted with scientists from the Far North Agricultural Research Institute, Norilsk, Russia; the U.S. Environmental Protection Agency; and the University of Oregon. In addition, EML also participates in the U.S. Interagency Arctic Policy Coordinating Committee (IARPC), and the International Arctic Monitoring Assessment Program (AMAP). Also, EML continues to pursue joint field studies with Russian scientists on environmental radiation measurement methodology.

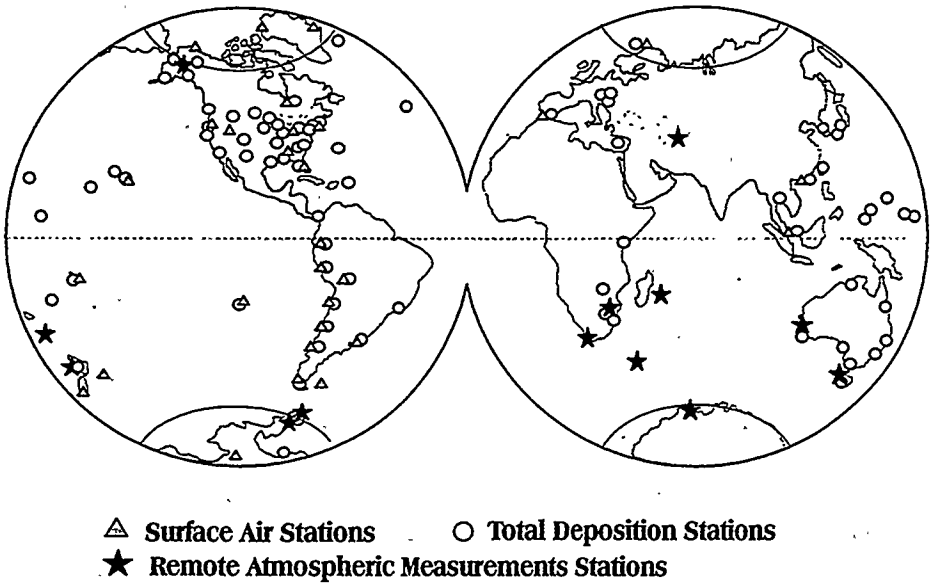

Figure 1.1 EML's global sampling network.

1.2 GLOBAL RADIONUCLIDE DEPOSITION STUDIES

Matthew Monetti, Karin M. Decker, Pamela Greenlaw, Sylvia Hulse, Catherine S. Klusek, William Rivera and Colin G. Sanderson

Since 1958, the intent of this program has been to study the global transport and fate of radionuclides released via atmospheric testing of nuclear weapons through the study of a radiologically important fission product, ^{90}Sr. Many factors controlling the global distribution of ^{90}Sr were identified through the operation of this program. Besides providing data useful for determining the radiological consequences of this global contamination, the fallout data is also valuable for studies using anthropogenic radionuclides as biogeochemical tracers of various processes. It has now been over 14 years since the last atmospheric test. During this period, ^{90}Sr was not detected in most of the samples analyzed. An exception was for many of the samples collected following the Chernobyl Accident in April of 1986 (Monetti and Larsen, 1991). At other times, detectable quantities of ^{90}Sr in the samples probably occurred due to resuspension of previously deposited material or minor localized releases of radioactivity from nuclear facilities.

At the end of 1994, this program, formally known as the EML Global Fallout Program, was in the final stages of a significant change. The new objective is to investigate the global deposition of a suite of radionuclides using gamma spectrometry instead of the previously focused on single fission product, ^{90}Sr. The radionuclides of particular interest include: naturally-produced isotopes, ^{7}Be and ^{210}Pb, and fission products, ^{137}Cs, ^{95}Zr and ^{144}Ce. It is expected that this change will expand the usefulness of the program, yet still maintain the original intent of the Global Fallout Program.

Gamma spectrometric measurements of fission products will enable us to identify accidental or intentional atmospheric releases of radioactivity. If releases should occur, then additional information can be obtained on the processes affecting the global transport and fate of these radionuclides. Follow-up studies could also be performed with these samples since the gamma analysis is nondestructive. The ^{7}Be and ^{210}Pb data are of special interest to researchers developing global circulation models. The deposition of these naturally-occurring radionuclides could be used to verify global circulation models. The data would also be available to calculate the source term and inventories of ^{210}Pb and ^{7}Be for researchers interested in using these radionuclides as biogeochemical tracers. Another advantage to changing this program is that the results will be better coupled to those found in SASP (see Summary No. 1.3) because gamma emitting radionuclides are measured in both programs. Comparison between these two programs can be used to study factors involved in the atmospheric removal of these radionuclides.

By the end of 1994, deposition samples were being collected at the 82 locations shown in Figure 1.1. There were no changes in the number or locations of these sites from the previous year. Monthly samples are collected at most of the sites using either an ion-exchange column, a stainless-steel pot or a polyethylene bucket. However, samples from the Australian sites and six sites managed by the United Kingdom's Atomic Energy Authority are collected on a quarterly basis. Along with the analytical change in the program there are also some changes in the processing of the samples. Now, all of the monthly collections, except for those collected at eight specific locations, will be composited into quarterly samples at EML. The samples collected at these specific sites will be analyzed as monthly samples in order to obtain more detailed results of the radionuclide (particularly the short-lived ones) deposition. The samples will be analyzed by gamma spectrometry, and then used for other studies or archived for potential future use. A new database will be started for the deposition of the gamma-emitting radionuclides. If fission products are detected, then an effort will be made to continue the previous 37 year old ^{90}Sr database. This can be accomplished either through direct measurements of ^{90}Sr or by calculating ^{90}Sr values based on isotopic ratios following the release. The data will be used to investigate temporal trends, total deposition and the global distribution of these radionuclides.

During 1994, additional studies were performed to verify earlier results reported last year which indicated the potential to use the sampling devices to collect the gamma-emitters of interest. The present results suggest that the samplers already in use in the program would be reliable for collecting the radionuclides of interest. The ^{210}Pb measurements may have high errors associated with them as a result of a high background relative to the total sample activity. Future modifications will attempt to address this situation. New procedures were developed during 1994 for the preparation of the samples for gamma spectrometric analysis. Some of the previously collected samples were processed and analyzed using these procedures. The new procedures will begin on a full scale early next year. Priority will be given to the 1995 samples, and the previously collected samples will be prepared and analyzed as time permits. A report is now in progress to present the results of ^{90}Sr in deposition during the years 1987 through 1990.

Reference

Monetti, M. A. and R. J. Larsen
"Worldwide Deposition of ^{90}Sr Through 1986"
USDOE Report EML-533, April (1991)

1.3 THE SURFACE AIR SAMPLING PROGRAM (SASP)

John Kada, Richard J. Larsen and
Colin G. Sanderson

SASP, initiated in 1957, is operated as a global network of high volume aerosol samplers at 45 sites around the world. Historically, the objective of this program has been to track on a global scale the dispersion of nuclear debris from the detonation of nuclear weapons. The program also has provided the capability of monitoring releases of radionuclides from other anthropogenic sources, e.g., the 1986 accident at the Chernobyl nuclear power reactor and the Tomsk-7 accident in 1993.

Measurement of naturally produced radionuclides, such as ^{210}Pb and ^{7}Be, in surface air has become an important aspect of the SASP program in recent years. ^{7}Be is a cosmogenic isotope produced predominantly in the upper troposphere and stratosphere, while ^{210}Pb is produced from decay of ^{222}Rn emanating from continental soils. Because their source terms are well known, the dispersion of ^{210}Pb and ^{7}Be through the atmosphere serves to trace atmospheric transport and aerosol scavenging processes. Modelers have therefore incorporated ^{210}Pb and ^{7}Be into chemical tracer transport components of their global transport models. The observed distributions of ^{210}Pb and ^{7}Be serve as a test of atmospheric transport and wet scavenging parameterizations in these models.

SASP provides the most extensive set of ^{210}Pb and ^{7}Be measurements available to the modeling community, and we plan to add aerosol sampling sites in Africa, Asia and the Arctic in collaboration with the Global Atmosphere Watch (GAW) program of the United Nations and with other agencies. We will also start measuring several other naturally produced radionuclides, including ^{35}S, ^{32}P, and ^{210}Bi, which will provide additional valuable tests of transport, scavenging, and chemical parameterizations used in chemical tracer transport models of GCMs.

Through SASP, EML also participates in the study of the atmospheric cycles of stable chemical species of biogeochemical interest. The University of Miami (UOM) analyzes air filter samples from SASP sites in remote regions as part of its research on global aerosol chemistry. Special emphasis is placed by UOM on analysis of sulfur and nitrogen species that may impact on aerosol-related climate processes. The ^{210}Pb and ^{7}Be measurements provided by SASP have played a role in the understanding of atmospheric cycles of these stable chemical species as well. Because the source regions of ^{210}Pb and ^{7}Be are well known, correlation (or lack thereof) between the stable chemical species measured by UOM and ^{210}Pb and ^{7}Be measured by SASP provides useful supplemental information helpful for understanding the atmospheric cycles of the stable chemical species (see Summary Nos. 1.4 and 1.5). A report will be published summarizing SASP data for the years 1990-1993 (Larsen et al., in press).

Reference

Larsen, R. J. Sanderson, C. G. and J. Kada
"EML Surface Air Sampling Program, 1990 - 1993 Data"
USDOE Report, in press

1.4 MEASUREMENT OF ^{35}S AT SASP SITES

John Kada

SASP surface air ^{210}Pb and ^{7}Be concentration data have proven valuable for testing transport and aerosol scavenging components of global chemical transport models. ^{210}Pb and ^{7}Be are not the only naturally produced radionuclides present in the atmosphere. They are simply the most readily measurable because both nuclides are gamma emitters, and therefore they can be measured by gamma spectroscopy. Several other naturally produced radionuclides exist in the atmosphere that could provide additional information about atmospheric chemistry and transport, and thus can provide data of value to chemical transport modelers.

One such nuclide is ^{35}S (t1/2 = 87 d), a cosmogenic isotope produced by the spallation of atmospheric nitrogen. The altitude and latitude dependence of the atmospheric source term of ^{35}S and ^{7}Be are identical, with one atom of ^{35}S produced per 55 atoms of ^{7}Be (Lal and Peters, 1967). While ^{7}Be exists in the atmosphere attached to submicron aerosol, ^{35}S is initially produced in the form of $^{35}SO_2$ gas, a fraction of which then oxidizes to form $^{35}SO_4$ aerosol. Simultaneous measurement of $^{35}SO_2$, $^{35}SO_4$, and ^{7}Be in the atmosphere thus provides information about the rates of chemical and physical processes which transform and remove SO_2 from the atmosphere, i.e. oxidation of SO_2 to SO_4, washout of SO_2 and SO_4, and dry deposition of SO_2 (Turekian and Tanaka, 1992).

The potential of ^{35}S to provide information about the dry deposition rate of SO_2 is particularly intriguing prospect. Because the factors influencing the dry deposition of SO_2 are so varied it has proven difficult to generalize results obtained from field and theoretical studies into an unequivocal characterization of SO_2 dry deposition on regional and seasonal scales (Erisman and Baldocchi, 1994). Yet such information is critical to developing accurate global chemical transport models of the atmospheric sulfur cycle because dry deposition is thought to be a significant sink of atmospheric SO_2, removing it from the atmosphere without forming climatically active SO_4 aerosol (Langner et al., 1992). Just as SASP surface air ^{210}Pb and ^{7}Be concentration data provides a test of atmospheric transport and wet scavenging of aerosol in global chemical tracer models, surface air $^{35}SO_4$ and $^{35}SO_2$ concentration data can be used to test parameterizations needed to accurately model the atmospheric sulfur cycle.

During 1994 we began discussions with a modeling group interested in incorporating ^{35}S into an global atmospheric sulfur cycle model. Our goal in the upcoming year is to begin to provide surface air ^{35}S data in support of such an effort. In the upcoming year we plan to begin a regular measurement of $^{35}SO_4$ concentrations in surface air at a subset of SASP sites representing different sulfur chemical climatologies. The measurements will be carried through an entire year to look for seasonal trends. We also will evaluate a multifilter cartridge method which would allow dual measurement of $^{35}SO_2$ and $^{35}SO_4$ at SASP sites using our standard high volume air sampling equipment. (see Summary Nos. 1.3 and 1.5).

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"Modeling Dry Deposition of SO_2"
Tellus, 46B, 159-171 (1994)

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Encyclopedia of Physics, vol. 46/2,
pp. 551-612, Springer, New York (1967)

Langner, J., et al.
"Anthropogenic Influence on the Distribution of Tropospheric Sulphate Aerosol"
Nature, 359, 712-716 (1992

Turekian, K. and N. Tanaka
"The use of Atmospheric Cosmogenic ^{35}S and ^{7}Be in Determining Depositional Fluxes of SO_2"
Geophysical Research Letters, 19, 1767-1770 (1992)

1.5 THE REMOTE ATMOSPHERIC MEASUREMENTS PROGRAM (RAMP)

Colin G. Sanderson, Norman Latner, S. Frederick Guggenheim, Vincent C. Negro, Scott Worms, Norman Chiu, Camille Marinetti, Steven Minick and Richard J. Larsen

RAMP was initiated in 1987 as an extension and modification of SASP (see Summary No. 1.3). In addition to having the goals of SASP, RAMP provides near real-time measurements of gamma-ray emitting radionuclides from remote or weathered-in regions around the world (Sanderson et al., 1994). Thus, radioisotopes having short half-lives, which normally would decay during the period between collection and analysis at EML, are easily measured. In addition, receiving data on a near real-time basis provides EML with a rapid response capability in the event of a nuclear accident.

In a collaborative effort with EML, the University of Miami also analyses selected RAMP filters for sulfur and nitrogen species which play an important role in aerosol chemistry and may also impact on aerosol-related climate processes. These studies continue to increase the understanding of the atmospheric aerosol which may have a significant impact on the cloud microphysics, precipitation chemistry, and the radiation balance of the entire Antarctic Region and the Southern Oceans.

To accomplish these objectives, RAMP sites (see Figure 1.1) are equipped with remote atmospheric measurements systems (RAMS) which measure the gamma-ray activity in air filter samples on site using either a sodium iodide detector or a mechanically-cooled germanium detector. The resulting spectra are: transmitted to the ARGOS communication system flown aboard the National Oceanic and Atmospheric Administration (NOAA) satellites or high throughput geostationary satellites, transferred to ground stations, and automatically recovered via a telephone link by EML's computer (see Figure 1.2). The spectra are then automatically reconstructed and analyzed at EML, and the data are available within 24 hours after field analysis. The analysis system automatically checks data validity and notifies responsible personnel of suspicious analysis results via voice and e-mail, and it maintains a complete current backup set of data and software at all times.

New system designs and installations:

--- In October 1994, a sodium iodide RAMS was installed near Fairbanks, Alaska. This system utilizes land telephone lines for two-way communications between the remote site and EML. Personnel at EML can now perform the same functions that site personnel perform.

--- A new, completely automatic RAMS was designed, developed, constructed and tested at EML (see Figure 1.3). Unattended operation of EML's RAMS is now possible for 1 month, if daily samples are collected, or up to 3 months, if samples are collected on a 3-day cycle. With this new system, the gamma-ray detector is an electrically cooled high-purity germanium diode coupled to a portable multichannel analyzer (MCA).

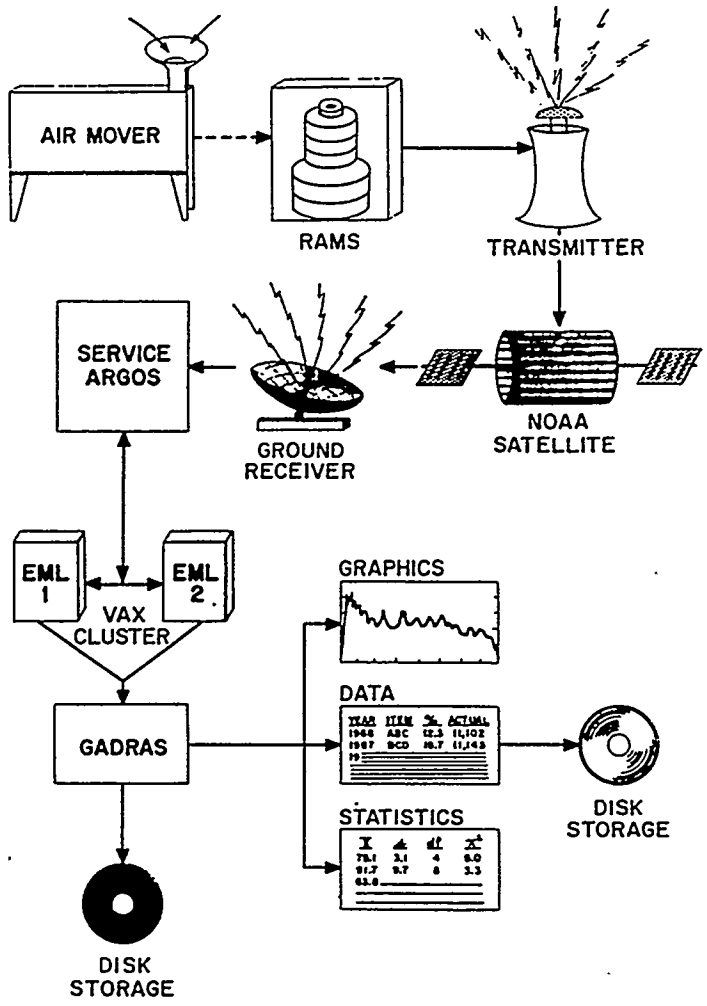

Figure 1.2 A schematic of the RAMP system.

A notebook computer is used to control the MCA, a "pick & place" robot system, and data flow. Sealed lead acid gel-cell batteries, trickle chargers and a transformer are used for continuous d.c. current operation. The transmitted spectra and onsite peak search analysis are reconstructed and reviewed at EML.

Future RAMP research will focus on high speed transmission of data for automatic collection and analysis. The use of INMARSAT-M satellites for two-way communications will provide EML with the ability to control the RAMS at remote locations and to recover gamma-ray data -instantaneously on demand where land telephones are either unavailable or unreliable.

Reference

Sanderson, C. G., N. Latner and R. J. Larsen "Environmental Gamma-ray Spectroscopy at Remote Sites with Satellite Data Transmission"
Nucl. Instrum. & Methods in Phys., A339, 271-277 (1994)

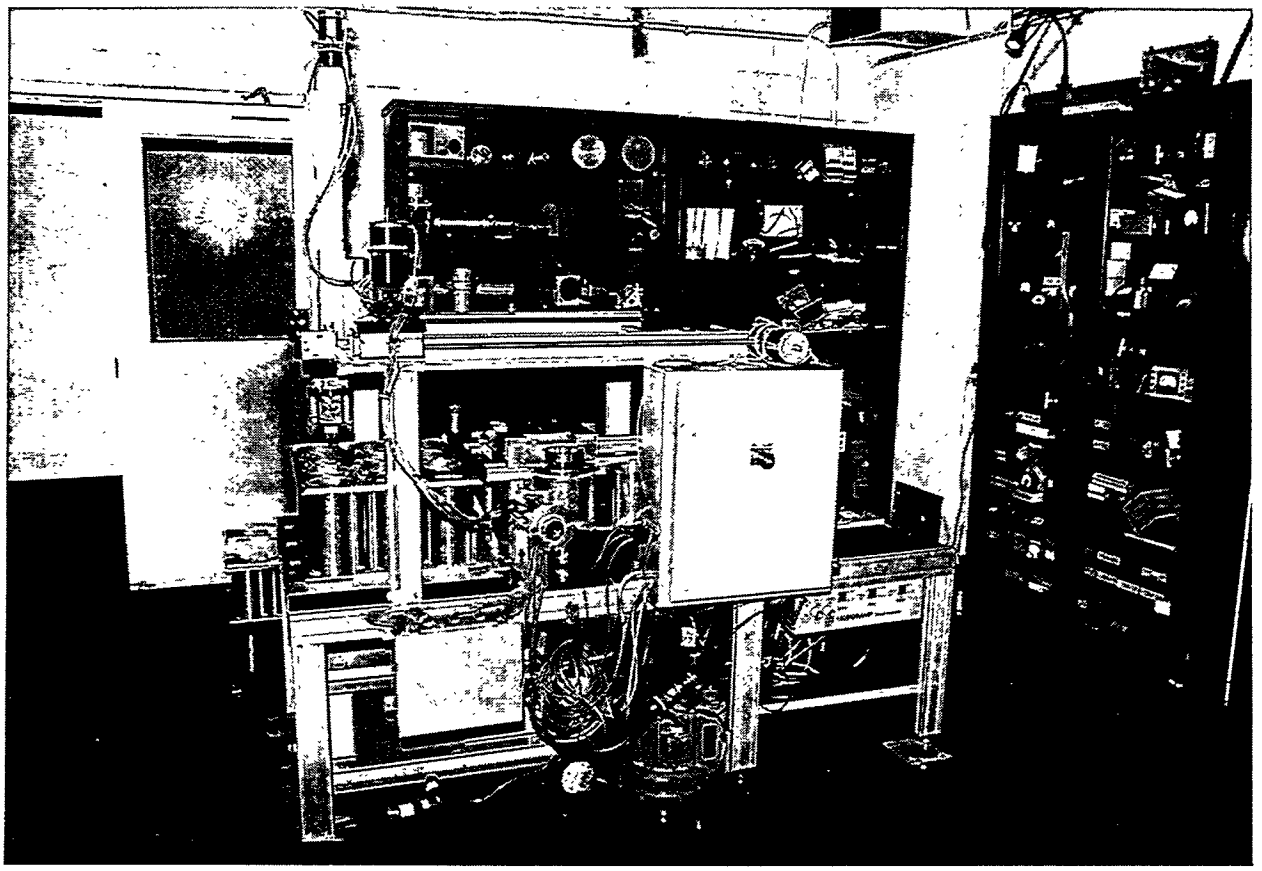

Figure 1.3 Automatic RAMPS system.

1.6 THE GLOBAL ATMOSPHERIC WATCH: EML's ROLE IN QUALITY ASSURANCE

Richard J. Larsen, Philip W. Krey and Merrill Heit

EML has been designated as the Center of Excellence for atmospheric radioactivity monitoring in GAW's Quality Assurance/Science Activity Center for the Americas (QA/SAC-Americas). In this new role, EML will initially provide direction to the GAW in the measurement of naturally occurring radionuclides, specifically, ^{7}Be, ^{210}Pb and radon progeny. QC issues, such as filter paper efficiency, gamma-ray spectrometric analyses and volume determination will also be addressed. Philip W. Krey, Acting EML Director, is a member of the Principals' Committee that will oversee this QA effort for the GAW.

GAW was created in June 1989 to enhance the World Meteorological Organization's (WMO) environmental data-gathering programs that began in the 1950s. The GAW integrates many existing monitoring and research studies that are being

conducted on chemical and physical properties of the atmosphere. The GAW also focuses on issues involving changes in the atmospheric concentration of greenhouse gases, changes in the ozone layer, long-range transport of pollutants, radionuclides in the atmosphere, acidity and toxicity of rain, as well as on the atmospheric burden of aerosols. The QA/SAC-Americas will play a leading role in defining data quality objectives and QA standards for the GAW.

1.7 ENVIRONMENTAL RADIATION MEASUREMENTS USING PRESSURIZED IONIZATION CHAMBERS

Peter Shebell, Kevin M. Miller, William Van Steveninck, Vincent C. Negro and Gladys A. Klemic

Routine monitoring of the penetrating component of the environmental radiation field (gamma plus cosmic) using pressurized ionization chambers is continuing at our regional baseline station in Chester, NJ. Additional monitoring is done at Princeton, NJ. This study is part of a long-term program whose goal is to document the natural variations as well as any anthropogenic perturbations in the natural background.

This year marked the first full year of monitoring with the new ionization chamber system at our Chester location. The system logged a total of 488,425 data points, yielding 92.9% coverage. For 2 minutes of every hour the system is recording an electrometer zero, therefore, the maximum yield is 96.67%. The system was down a total of 13.66 days in 1994 due to maintenance and equipment problems.

Improvements in the data storage and analysis program included performance tests on the CMOS memory cards, as well as safeguards against the accidental loss of data. Other work on the new system included the determination of its angular response.

The average dose rate in air at the Chester site in 1994 was 120.5 nGy h^{-1}. The maximum dose rate (averaged over 1 minute) was 182.3 nGy h^{-1} recorded on 11/10/94 at 1:06. The minimum dose rate was 65.53 nGy h^{-1} recorded on 3/7/94 at 15:59. These variations are consistent with those normally found at this location.

The Princeton location also logged its first full year of monitoring. The system recorded 409,898 dose rate values for a yield of 78%. The system was down 68.19 days in 1994 due to maintenance and equipment problems. Failures in the CMOS memory cards were the reason for the increase in down time. In one continuous run, the system successfully recorded 59,311 dose rate values, or 42.61 days of data. This represents the longest unattended field exposure for an EML monitoring ionization chamber system.

The average dose rate at the Princeton location in 1994 was 59.74 nGy h^{-1}. The maximum dose rate was 193.5 nGy h^{-1} recorded on 5/18/94 at 19:30. The minimum dose rate was 36.62 nGy h^{-1} recorded on 2/16/94 at 16:25. This site is at the property line of the Princeton Plasma Physics Laboratory. The significant increase in dose rate over the annual average is a result of high-power deuterium-tritium experiments at the TFTR (see Summary No. 2.2).

Analysis of our environmental radiation monitoring database began this year, which includes a statistical profile of the entire 18 years of daily dose rate measurements. Similar profiles of shorter-time periods to document monthly, quarterly, yearly, as well as seasonal variations will also be provided. The results of this analysis will be presented at the Natural Radiation Environment VI Symposium to be held in 1995.

1. 8 COMMISSION OF EUROPEAN COMMUNITIES (CEC) INTERCALIBRATION EXPERIMENT

Gladys Klemic, Peter Shebell and Kevin M. Miller

EML was invited to represent the U. S. in an international intercalibration experiment sponsored by the CEC in June of 1994. The focus of the 2-week project was the measurement of environmental radiation, with a goal toward assuring accurate monitoring of the global environment. Such intercomparisons were initiated among European countries in 1984; this was the first time the U.S. has participated. Other countries participating were Czech Republic, Denmark, Germany, Hungary, Poland, Spain, and the United Kingdom.

EML used three instruments during the intercomparison: a high purity germanium detector for gamma spectrometry, a small (portable) pressurized ionization chamber for environmental radiation, and EML's newly redesigned pressurized ionization chamber that is used for monitoring. Measurements began at the Physikalisch Technische Bundesanstalt (PTB) in Braunschweig, Germany, using a 6 MeV photon beam to irradiate the detectors. This is the first time EML's instruments were calibrated in such a high-energy field. Additional measurements were performed at the PTB's ultra low level calibration facility in a salt mine 900 m below ground, where the background radiation is about 100 times less than that at sea level, on the order of 1 nGy h^{-1} (~ 0.1 μR h^{-1}). This facility provided an opportunity to check the linearity of instruments using five low level

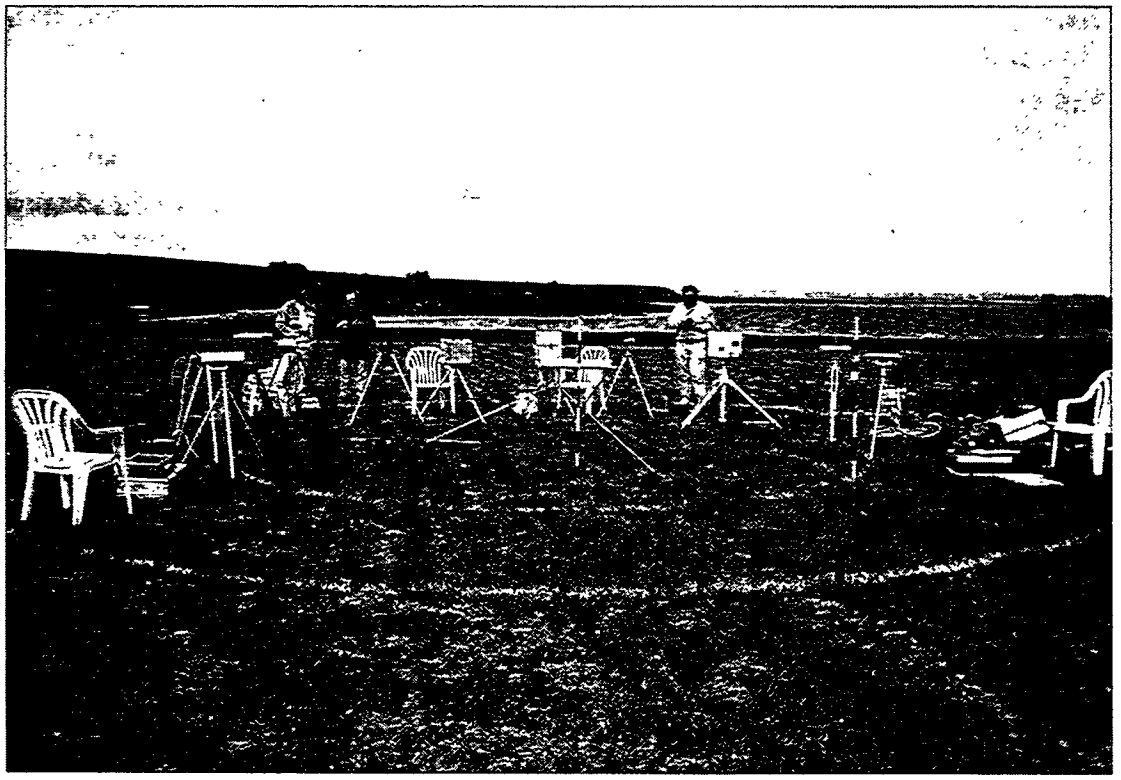

Figure 1.4 Free field calibration of various instruments at the Risø National Laboratory environmental testing station in Roskilde, Denmark.

(~ 10 - 150 nGy h^{-1}) ^{137}Cs calibration sources, as well as to determine the inherent background of the instruments.

The following week, measurements continued at the Risø National Laboratory in Roskilde, Denmark (see Figure 1.4). Free field calibrations were performed outdoors using three different sources (^{60}Co, ^{137}Cs and ^{226}Ra) for various energy photons. Natural background radiation was also measured at the field site. Shadow shield calibrations were performed in a laboratory using the ^{137}Cs source. Measurements of cosmic radiation were performed on a wooden pier over the Roskilde fjord, and also on a ship that cruised on the fjord for several hours. The results from this intercalibration will be included in a progress report to the CEC, and will be submitted to a peer-reviewed journal for publication in collaboration with the other participants.

EML's participation in this project has lead to other collaborations. The PTB scientists accepted an invitation to visit EML and National Institute of Standards and Technology (NIST) this September to present a seminar on their facility, and also to discuss the possibility of establishing similar environmental calibration standards at NIST. A representative from EML was invited to sit in as a corresponding member on the newly established working group on "Environmental Radiation Monitoring" within the European Radiation Dosimetry Group (EURADOS).

1.9 ENVIRONMENTAL THERMOLUMINESCENCE DOSIMETRY

Gladys Klemic, Peter Shebell and Kevin M. Miller

EML's thermoluminescence dosimetry (TLD) program encompasses three complementary areas of research directed toward environmental applications of TLDs: (1) studies related to routine monitoring; (2) planning and implementation of international intercomparisons; and (3) contributions to the development of national standards. In addition, other TLD research focuses on investigations of the use of TLDs in mixed radiation fields with potential applications in personnel dosimetry (see Summary No. 1.9).

Routine Monitoring Research. To provide a framework for testing new equipment and techniques, EML maintains two outdoor monitoring sites where TLDs are deployed next to continuously monitoring pressurized ionization chambers (see Summary 1.7). Methods and materials that have been established through a 20 year residential monitoring program are still used to provide an integrated measurement of gamma- and cosmic-ray natural background, while new phosphors and analysis methods are being tested. Two new readers now supplement the EML designed reader that has been in use since 1974. An automated hot gas reader is used for routine measurements and large scale applications, including quality control (QC) for the international intercomparisons and batch testing of chips. A commercially available manual (single chip) pan heating reader is being used for research applications, including glow curve deconvolution. Improved uncertainty analysis and QC checks within EML's TLD analysis software and other developments in EML's standard procedures were described in a major revision of the Procedures Manual written in 1994.

International Intercomparisons. The International intercomparisons of environmental dosimeters were initiated in 1974 to assess the performance of passive, integrating detectors in the measurement of environmental radiation and to identify and investigate special problems associated with such measurements. These intercomparisons have become a popular means for scientists to measure their techniques along side those of their peers worldwide. The program is

voluntary and the results are reported without identifying individual participants. In 1994, analysis of the results of the 10th intercomparison was completed for publication (Klemic *et al.*, in press). One hundred and two scientists from 27 countries participated in this intercomparison, which included a 3 month outdoor field exposure and irradiations with a laboratory ^{137}Cs source at the Idaho National Engineering Laboratory (see Figures 1.5 and 1.6).

Planning of the 11th intercomparison began with the establishment of a new collaboration with Brookhaven National Laboratory (BNL) and the exploration of potential field sites at BNL. Further developments included preliminary designs for a possible special experiment to include a low energy photon exposure, which could be a concern for power plant monitoring should an accident occur, as learned from the accident at Three Mile Island.

National Standards Work. In November 1994, EML participated in meetings of the ANSI working groups N13.29 and N13.37. ANSI Standard N13.37 is expected to replace an earlier document (N545) as the type testing standard for environmental TLD systems. ANSI Standard 13.29 will establish performance criteria for environmental dosimetry processors and will be analogous to a similar standard for personnel dosimetry (N13.11), which is currently the basis for the DOE's Laboratory Accreditation Program (DOELAP). The analysis of the 10th intercomparison results proved useful in helping to set tolerance levels for these criteria. Special experi-

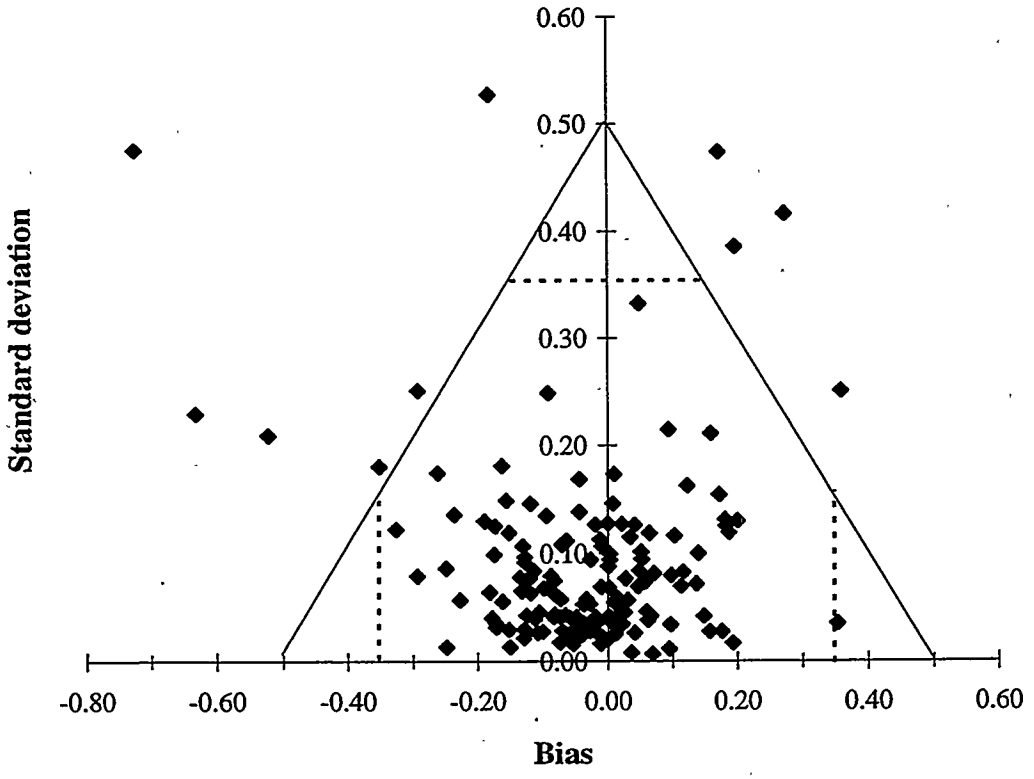

Figure 1.5 Results of the 10th International Intercomparison of Environmental Dosimeters, illustrating the performance as defined in ANSI standard N13.11, where each point represents one dosimeter set, with the bias plotted on the x-axis and the standard deviation on the y-axis. Triangle encloses those within proposed ANSI N13.29 tolerance level of 0.5. This demonstrates how the intercomparisons provide useful data for national standards work.

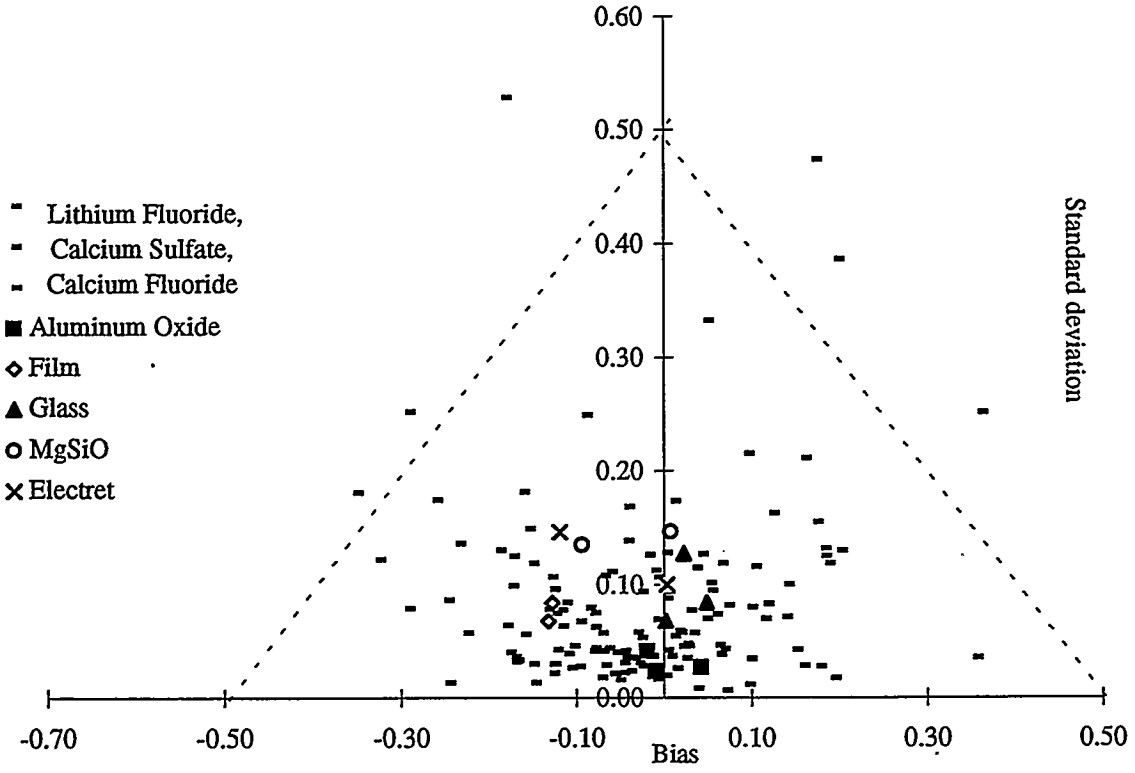

Figure 1.6 Same data, as in Figure 1.5, emphasizing the performance of some of the newer TL phosphors and non-TL dosimeters compared to the three most widely used TLDs.

ments are being planned for the 11th intercomparison to address some other issues relevant to these standards. Collaborations established from participation in CEC sponsored research (see Summary No. 1.8) also proved useful to put U.S. standards in context with current trends in international standardization being developed in Europe.

Reference

Klemic, G., J. Shobe, T. Gesell, and P. Shebell "Results of the Tenth International Intercomparison of Environmental Dosemeters" Radiation Protection Dosimetry, 58, 133-142 (in press)

1.10 PLUTONIUM ISOTOPES IN SOILS AT THE IDAHO NATIONAL ENGINEERING LABORATORY (INEL)

Thomas M. Beasley

$^{240}Pu/^{239}Pu$ isotopic ratio measurements are being used to delineate the dispersion of fuel reprocessing plutonium wastes at the INEL. Figure 1.7 shows soil coring sites directionally away from the Radioactive Waste Management Complex (RWMC). Transuranic wastes from Rocky Flats, CO, have been stored at this facility for several decades. Accidental releases of this material have occurred and plutonium has been dispersed to the surrounding area by wind-driven resuspension.

Soils to the east of the INEL, in Montana and Wyoming, show the presence of fallout from the Nevada Test Site where $^{240}Pu/^{239}Pu$ ratios are comparable to those of fuel reprocessing. However, soils within 100 km of the INEL site boundaries (in both westerly and easterly directions), and over much of the site itself, have plutonium isotopic signatures that are entirely consistent with integrated global fallout ratios.

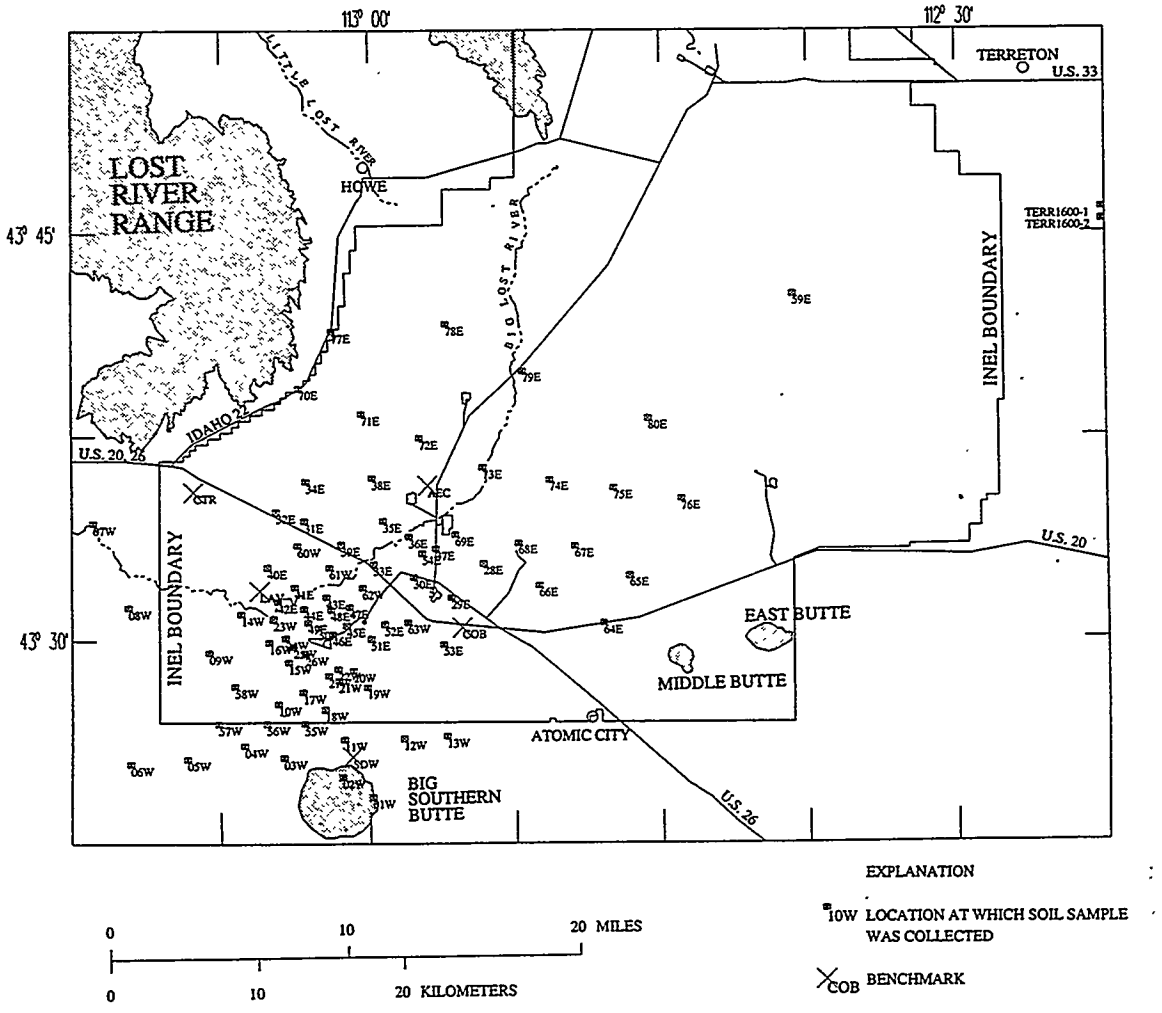

Figure 1.7 Soil coring sites for isotopic plutonium measurements at the INEL Positions are fixed with a Ground Positioning System operated by the U.S. Geological Survey Project Office.

Results from cores near the southern site boundary (56W, 57W, 10W and 15W) indicate that plutonium released from the RWMC has been transported in a south-westerly direction to these sites; there is no indication that RWMC-derived material has been transported to greater distances in this direction. In the north-easterly direction, RWMC-derived material has been detected at coring site 38 E and at sites closer to the RWMC. The trajectory of the dispersion is consistent with the directional flow of winds across the INEL which change diurnally. Sampling to greater distances in the north-easterly direction is required because of the higher velocities of winds originating from western Idaho.

It is anticipated that by the end of calendar year 1995, sample analyses will be completed and isopleths of plutonium dispersion away from the RWMC can be constructed.

1.11 CONTAMINANT DEPOSITION STUDIES IN ARCTIC ALASKA

Matthew A. Monetti, Yulin L. Tan, John Kada, Kevin M. Miller and Ada Kong

Although the Arctic is believed to be one of the most remote places on earth, there is a potential that it could be impacted by anthropogenic activities. Observations of a seasonal phenomenon known as "Arctic haze" have been reported for the past 40 y. As the Arctic mass expands during the Northern Hemisphere's winter, the atmospheric circulation patterns change. This change potentially allows pollutants to be transported into the Arctic from lower latitudes, particularly from Eurasia.

Our studies began in 1991 through scientific collaboration in the EPA's Arctic Contaminants Research Project. EML's interest is in documenting and interpreting the presence of combustion produced, potentially carcinogenic organic compounds, such as polycyclic aromatic hydrocarbons (PAH) and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/F) and radionuclides (both natural and anthropogenic). Our main approach in this study is to investigate the chronological record of these contaminants that can revealed through the use of lake sediments. Sediment cores from nine lakes, stretching over 650 km across the Alaskan Arctic, have been successfully collected during this study. These samples should allow us to adequately address and interpret the variations in contamination across this vast Arctic wilderness.

During 1994, the final year of field sampling activities for this project, lake sediment cores were collected from three additional lakes in the central North Slope region of the Alaskan Arctic. Two of the sediment cores were counted by gamma spectrometric methods. The 1994 results were similar to those found in previously counted sediment cores.

A summary of all the ^{137}Cs and excess ^{210}Pb data from these Arctic studies to date is shown in Table 1.1. This radionuclide data is particularly useful for dating the sediment cores. The ^{137}Cs and excess ^{210}Pb is present in only the upper few centimeters of the cores since the sedimentation rate is low in these Arctic systems. The ^{137}Cs inventories found in these cores ranged from 560 to 2700 Bq m^{-2}, and are generally higher than the expected inventories due to sediment focusing. This radionuclide data will be prepared for publication as a journal article.

Results of the PAH from Lake Schrader sediments in the Arctic National Wildlife Refuge are now available. In general, the values for the parental PAH are slightly lower than the concentrations found in the sediments from a sub-Arctic Alaskan lake, Wonder Lake (Tan et al., 1993). There is no apparent temporal trend to the concentrations found in the Lake Schrader core, so there is little indication that this region has been affected through the deposition of PAH resulting from the "Arctic Haze". The PAH results from lake Schrader will also be published, and will add to the understanding of the contamination of the Arctic environment. (see Summary No. 1.13).

Reference

Tan, Y. L., A. Kong, M. A. Monetti, B. Lau, C. Gubala and D. Landers
"Polycyclic Aromatic Hydrocarbons and Polychlorinated Dibenzo-P-Dioxins and DibenzoFurans in Sediment from a Sub-Arctic Lake in Alaska"
in: H. Fiedler, H. Frank, O. Hutzinger, W. Parzefall, A. Riss, S. Safe (Editors)
13th International Symposium on Chlorinated Dioxins and Related Compounds, September (1993)

TABLE 1.1

SUMMARY OF ARCTIC LAKE GAMMA RADIONUCLIDE DATA

Lake - Core	^{137}Cs Peak (cm)	^{137}Cs Penetration (cm)	^{137}Cs inventory (Bq m^{-2})	^{210}Pb Penetration (cm)	^{210}Pb inventory (Bq m^{-2})	Sedimentation Rate (mg cm^{-2} y^{-1})
Wonder - 1	Surface	7	560	6	3140	8
Wonder - 2	Surface	8	1300	5	5950	5
Schrader - 1	4	10	2570	> 11	> 4800	86
Schrader - 2	3	10	-	-	-	-
Schrader - 3	3	10	-	-	-	-
Schrader - 10	Surface	4	900	6	1090	27
76 - 3	2	7	-	-	-	-
Feniak - 1	3	7	2770	-	-	28
Feniak - 2	2	7	2710	7	1900	23
Feniak - 3	3	7	2260	-	-	28
Desperation - 1	5	8	2460	> 9	> 2320	73
Chandler - 1	3	5	1720	> 7	> 1720	24
Chandler - 2	3	5	1680	-	-	22
Nyagome - 1	3	6	2180	>7	> 2970	61
Khutudaturku - 1	Surface	5	654	5	750	19
Elusive - 2	2	6	1580	7	1920	22
Toolik - 2	Surface	> 7	> 1440	> 7	> 1780	9

1.12 BIOGENIC PAH IN AN ALASKAN ARCTIC LAKE SEDIMENT

Yulin L. Tan, Ada Kong and
Matthew Monetti

A 30-cm sediment core from Lake Schrader in Alaska, dating back about 270 years, was analyzed (see Summary No. 1.12). The core was segmented and dated for chronological pollutant deposition studies. The dated sediment segments were analyzed for three- to six-ring PAH by gas chromatography/mass spectrometry following the procedures developed at EML (Tan et al., 1993). The PAH composition profiles as revealed by the gas chromatograms are distinctively different from the PAH composition profiles of environmental samples that are contaminated by anthropogenic activities. The major PAH in polluted environmental samples consists of over a dozen parental PAH with typical composition profiles indicating combustion origins. In the Schrader Lake sediment, the presence of parental PAH were found to be overwhelmed by a group of biogenic PAH, leading to totally different PAH composition profiles.

The major biogenic PAH from diageneses of natural biological products found in the Schrader Lake sediment are phenanthrene, alkylated phenanthrenes, alkylated tetra- and octa-hydrochrysenes, and perylene. Their original biological precursors were traced by their preserved carbon skeletons and were found to be diterpenoid and triterpenoid biolipids and perylene-quinone pigments. The depth profiles of these PAH reflect the rate of the diagenetic processes.

Diagenesis is the first stage of transformation which carries the original biological lipids toward the maturarion to fossil fuel formation. Therefore, biogenic PAH entrained in petroleum can be useful biogenic markers in petroleum geochemistry. The results of this study will be presented at the 15th International Symposium on Polycyclic Aromatic Compounds to be held in Belgirate, Italy in September of 1995.

Reference

Tan Y. L., A. Kong and Y. O. Chiu
"Sample Preparation for Analyzing Polycyclic Aromatic Hydrocarbons and Polychlorinated Dibenzo-p-dioxins and Dibenzofurans in Sediment by Gas Chromatography/Mass Spectrometry"
Estuaries, 16, 427-32 (1993)

1.13 RADIONUCLIDES THE ARCTIC OCEAN BASIN

Thomas M. Beasley

During 1994, a substantial number of ^{129}I, ^{237}Np, and isotopic Plutonium measurements were made in support of the Office of Naval Research's ANWAP Program (Arctic Waste Assessment Program). This study is being conducted in collaboration with scientists from ORNL, ANL, LANL, and the IsoTrace Laboratory of the University of Toronto. It is part of a larger effort to investigate radioactivity releases by the former Soviet Union to the Arctic Ocean Basin.

From data collected to date, we have concluded the following: ^{129}I released from the fuel reprocessing facilities at Sellafield (U.K.) and La Hague (France) have been advected through the Barents and Kara Seas to the Laptev Sea (Figure 1.8). Iodine-129 atom concentrations in surface waters of all three water bodies are similar (~ 12-15 x 10^8 atoms L^{-1}) and exceed expected "background" levels (~ 0.1-0.2 x 10^8 atoms L^{-1}) by an order of magnitude. Iodine-129 concentrations in the Siberian Coastal Current (Chukchi Sea) show no influence of fuel reprocessing input; atom concentrations are essentially background (0.2 x 10^8 atoms L^{-1}). There is evidence that fuel reprocessing ^{129}I has found it's way to the Canadian Basin near the Alaskan coast (Figure 1.9). Subsurface maxima in iodine concentrations are seen at depths of 200-400 m and reach levels of 3-4 x 10^8 atoms L^{-1}, and generally exceed surface water values at the same site by an order of magnitude. The origin of these labeled waters may be the continental shelf and

Iodine-129 atom concentrations
(10^8 atoms liter^{-1})

A&B	15±4	(n = 36)
C	11±1	(n = 11)
D	0.20±0.08	(n = 14)
E	0.3 - 4.7	(n = 19)

Fuel reprocessing Pu measured at sites C and E

Figure 1.8 Arctic Ocean Basin sampling sites.

slope of Kara, Barents and Laptev Seas where cold, saline waters form, sink, and then spread to the interior of the Arctic Ocean basins. We are presently analyzing a suite of samples from a trans-Arctic cruise (funded by the National Science Foundation) that will further characterize the extent to which ^{129}I released from European fuel reprocessing facilities has been distributed within the Arctic Ocean basin.

We have confirmed that fuel reprocessing plutonium is present in sediments of the Canadian basin (Figure 1.9). Fuel reprocessing plutonium typically exhibits $^{240}Pu/^{239}Pu$ isotopic ratios between 0.05 to 0.06. Integrated global fallout ratios for the same isotopes at the latitudes of the Arctic Ocean typically range between 0.18 and 0.19. For coring site 16, greater than 60% of the plutonium in the surface sediments has its origin as fuel reprocessing. Using $^{241}Pu/^{239}Pu$ ratios, it is possible to "date" the production time of the fuel reprocessing material once corrections have been made for the global fallout contribution to the plutonium measured. For the Canadian Basin material, this date corresponds closely to 1957, the time of a substantial, accidental release of radioactivity from the Mayak Complex in Cheliabinsk Province in the former Soviet Union (Trabalka et al., 1980). The absence of elevated levels of ^{238}Pu in sediments of the Barents and Kara Seas argues against the import of Sellafield-derived plutonium (Kershaw et al., 1990) as a source for the fuel reprocessing plutonium measured in the Canadian Basin. A suite of surface sediment samples, collected between the Bering Straits and the North Pole, are presently being analyzed for their isotopic plutonium content to further map the distribution of fuel reprocessing plutonium throughout the Arctic Ocean basins.

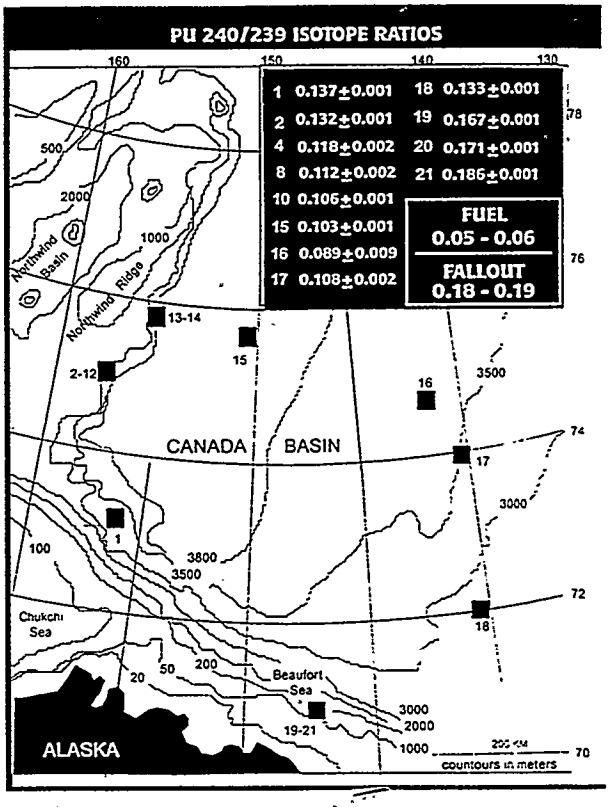

Figure 1.9 Isotopic plutonium in sediments collected from the Canadian Basin. Typical weapons-grade material and integrated fallout ratios are shown for comparison.

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1.14 OB RIVER/ESTUARY STUDY OF 1994

Matthew Monetti, Karin Decker,
William Rivera and
Colin G. Sanderson

As a result of accidents and testing at nuclear facilities such as Chelyibinsk, Tomsk and Semipalatinsk in the former Soviet Union, enormous quantities of radionuclides have been introduced into the Ob River watershed (Trabalka et al., 1980; Aarkrog et al., 1992; Larsen et al., 1994). Although there have been attempts to restrain this material so that it does not enter the Ob River, it is not unreasonable to suspect that the River may be a significant source to the Arctic.

During the summer of 1994, EML was invited by Dr. Hugh Livingstone and Dr. Fred Sayles of the Woods Hole Oceanographic Institute to participate in a project investigating the Ob River as a source of radionuclides to the Arctic. This involved EML's assistance with the field preparation, sampling activities and sample analyses. The sampling objective was to obtain sediment cores from the seasonally flooded region of the Ob River delta and estuary that would reveal the past history of radionuclide contamination. The samples were collected using equipment designed specifically for the purpose of the operation. Cores were retrieved from 15 stations during a cruise on a Russian fishing vessel along nearly 1600 km of the river's lowest reaches. A minimum of two cores were obtained at each location. The cores ranged from 30 to 120 cm in length. One core was sectioned by intervals of 1 cm to 5 cm on board the ship, and the other was left intact for further consideration on the best approach for sectioning.

Initial gamma radionuclide results on a couple of the cores indicate that these cores will be useful for investigating the historic record of radionuclide contamination along the lower portions of the river. These results suggest that the ^{137}Cs record is indicative of a source dominated by radioactive fallout, but further analyses are required for verification purpose. Additional analyses will be performed on these samples by the Woods Hole Oceanographic Institute group, and EML will provide additional assistance with the gamma spectrometric analyses as necessary. EML may be involved in a future excursion into regions further upstream on the Ob River in an attempt to determine the downstream extent of the suspected radionuclide contamination.

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2 Radiation Transport and Dosimetry

2.1 OVERVIEW

Paul Goldhagen

In the Radiation Transport and Dosimetry program, analytical and experimental techniques are applied to determine the basic physical properties of radiation fields in the workplace and in the environment. Projects within this program are often initiated in response to requests to properly evaluate occupational radiation exposure problems at DOE facilities or to provide critical exposure assessments for DOE or other agencies. These situations usually involve complex radiation fields containing neutrons and/or high-energy particles for which normal dosimetric techniques are either inadequate or questionable in the absence of adequate validation. We develop, use and provide reliable experimental and theoretical/calculational tools to evaluate the nature, magnitude, and potential health consequences of human exposure to such ionizing radiation. To do so, interactions are maintained with other advanced dosimetry research programs in the U.S. and in many other countries. Progress in this program contributes toward establishing a viable risk-based system for radiation protection, one that will have a beneficial impact on the conduct of DOE operations.

The evaluation, application, and development of computer codes for radiation transport (the propagation of radiation, including its interactions with matter) and for spectral unfolding continue to be an important part of this program. Using such codes, calculations are performed to: determine radiation fields from instrument readings, relate measured fields to the properties and distributions of radiation sources, and interpret the measurements in terms of dose to man. In many cases, calculations provide the basic information on radiation field properties with critical measurements providing validation.

The experimental part of this program has emphasized neutron dosimetry in recent years because of the continuing difficulties associated with making reliable assessments of neutron fields and exposures at DOE nuclear facilities and elsewhere. The conservatism introduced into the assessment of neutron dose equivalents resulting from the uncertainties of measurements affects the efficiency and cost of DOE facility operations. Because of the experience gained and the exper-

tise developed at EML in this field, notably with multisphere neutron spectrometer systems, the Laboratory has responded to a number of requests to provide reference neutron energy spectra at critical locations in or near nuclear facilities and to assess the neutron component and its contributions to dose in complex radiation fields. Two examples of this are ongoing projects in 1994, one at the Princeton Tokamak Fusion Test Reactor (TFTR), and the other at the Army Pulse Radiation Facility (APRF). The latter study was performed at the request of the Defense Nuclear Agency and is part of the broad effort to resolve the discrepancy between measured and calculated thermal neutron activation at Hiroshima, one of the most important unsolved problems in radiation dosimetry.

Another aspect of the experimental program has been the development of improved and new methods for neutron dosimetry. The energy response function matrix of the multisphere neutron spectrometer used in the measurements at the TFTR and the APRF has been recalculated, which is likely to substantially improve the accuracy and reliability of the inferred neutron energy spectra. A new multisphere neutron spectrometer has been built which is more portable and rugged, and has higher neutron sensitivity and wider energy response than our earlier spectrometers. The new dosimetry systems, which are based on Al_2O_3 thermoluminescent detectors (TLDs) and were used at the TFTR and the APRF, appear to be significant advances in personnel dosimetry and in the passive measurement of thermal neutrons. Measurements and calculations to improve our understanding of the response of various TLDs to neutrons are described along with the development of a portable TLD reader.

Over the past several years, there has been an increasing awareness that aircraft crews receive one of the highest average dose equivalents of any occupationally exposed group. The National Aeronautics and Space Administration (NASA) is now considering designs for future high-speed high-altitude civil aircraft in which crews would be exposed to significantly higher levels of radiation. In collaboration with NASA's Langley Research Center, EML scientists have examined the radiation safety aspects of such future high-altitude commercial aircraft. Among several areas needing further research, it was found that there are large uncertainties (plus or minus a factor of 2) in our knowledge of the physical fields for high-energy neutrons and multicharged ions, and that neutrons can cause up to half or more of the biological damage. Neutrons also make a large and not accurately known contribution to the dose equivalents received by present-day airplane crews. Because of EML's expertise in neutron spectrometry, the Royal Military College of Canada and the Defense Research Establishment, Ottawa, invited us to join them in performing detailed measurements of the cosmic radiation field aboard dedicated flights of Canadian Forces aircraft. Much of the calculational and theoretical effort this year has also been directed toward cosmic radiation at high altitudes. In collaboration with the NASA Langley Research Center, analytical methods developed there were applied to calculate dosimetric quantities as a function of depth in tissue-equivalent materials for high-energy multicharged ions. A Monte Carlo particle transport code incorporating an intranuclear cascade plus evaporation model of nuclear fragmentation is being extended to higher energies in order to make it useful for cosmic-ray transport at high altitudes.

Although this program primarily involves research in support of radiation protection activities at DOE and other nuclear facilities, it also provides the basic scientific information needed for the development of the next generation of radiation protection instrumentation and methods. Thus, this research, while finding direct

application to current practical problems of concern to DOE, also has a broad generic value to the radiation protection community and is likely to facilitate the development of improved risk estimates for high-energy and high-LET radiation exposures.

Much of the work in this program involves varying degrees of collaboration with other research and radiation protection groups. Among them, in 1994, have been the Princeton Plasma Physics Laboratory, APRF, Science Applications International Corporation, NASA Langley Research Center, Los Alamos National Laboratory, Oak Ridge National Laboratory, the Royal Military College of Canada, the Defense Research Establishment, Ottawa, and the Mexican National Institute of Nuclear Studies.

2.2 MEASUREMENT OF RADIATION FIELDS NEAR THE PRINCETON TOKAMAK FUSION TEST REACTOR (TFTR)

Paul Goldhagen, Peter Shebell, Nestor Azziz, William Van Steveninck, Alfred Cavallo and Ferenc Hajnal

In 1994 the TFTR of the Princeton Plasma Physics Laboratory (PPPL) produced record-breaking pulses of fusion power using the deuterium-tritium (D-T) reaction, including a 10 MW pulse on November 2 (Scott et al., 1994; Associated Press, 1994). This achievement followed years of development and testing using the lower energy deuterium-deuterium (D-D) reaction. Since 1990, in collaboration with Henry Kugel of PPPL, we have been measuring neutron and gamma radiation field quantities at designated locations around the TFTR (Kugel et al., 1994a, b).

The EML measurements were originally undertaken at the request of PPPL to calibrate measurements by TFTR staff, provide benchmark data for shielding calculations, and assure that radiation protection limits would not be exceeded when D-T operation began. The TFTR shielding design objective for the PPPL property lines is to limit the dose equivalent to 0.1 mSv y^{-1} from all sources and pathways. Our early D-D measurements showed that the TFTR shielding would be adequate to meet this objective during planned D-T operations yielding $1x10^{21}$ D-T neutrons per year.

The instrumentation for measurements outside the Test Cell (the TFTR shielded enclosure) includes a multisphere neutron spectrometer, several pressurized ionization chambers (PICs), and a high-purity intrinsic germanium (HPGe) gamma-ray spectrometer. The neutron and gamma spectrometers and one of the PICs incorporate computer-controlled gated electronics to allow analysis of individual TFTR fusion pulses (~1 duration, 7 to 15 min apart), and these instruments are used together for comprehensive measurements. In 1994, comprehensive measurements were performed 120 m northeast of the center of TFTR, and additional continuous PIC gamma dose measurements were taken at other locations, including the nearest property line (see Summary No. 1.8).

We have already taken enough data to determine the neutron and gamma-ray spectra near and far from the shield walls for D-T fusion, but we are continuing our measurements in order to characterize the change in the radiation field as the fraction of D-T fusion varies from near 1% (D-D operation) to nearly 100% (full-power D-T operation), an important consideration in the design of future fusion reactors and their shielding. Figure 2.1 shows preliminary results from the HPGe gamma spectrometer for 32 fusion pulses with a trace of tritium in the plasma, such that the ratio of D-D to D-T neutrons was 1.25.

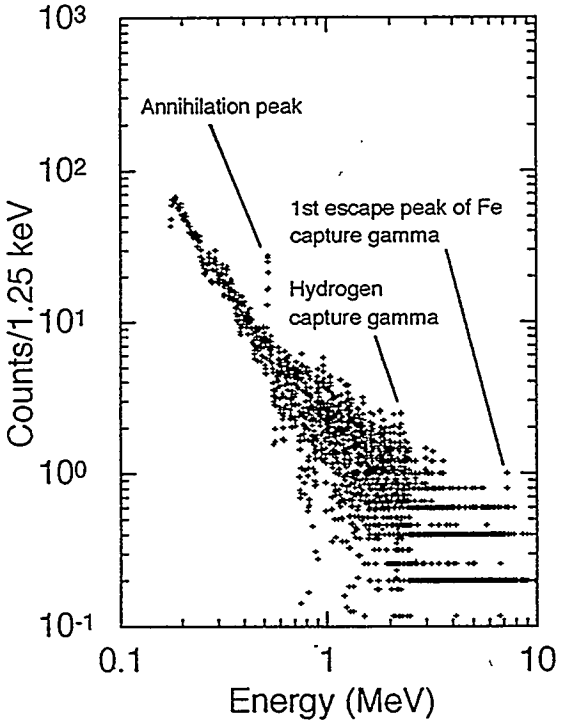

Figure 2.1 Composite Ge gamma detector spectrum taken 120 m northeastward of the TFTR during 32 fusion pulses having a slight tritium admixture (D-D/D-T neutrons = 1.25). Indicated are the first escape peak of iron (7.120 and 7.134 MeV) and the full energy absorption for hydrogen (2.223 MeV).

In addition to measurements outside the Test Cell, an experiment was carried out using TLDs to determine the ratio of gamma rays to neutrons (mainly thermal) inside the Test Cell during D-D and D-T fusion. Earlier studies at EML (Azziz and Azorin, 1993; Azziz et al., 1994) found that the insensitivity to neutrons of Al_2O_3:C TLDs, compared to their high gamma sensitivity, can be utilized in determining the ratio of gammas to thermal neutrons in mixed (n,g) fields. The technique makes use of bare TLDs and TLDs covered with cadmium, which emits gamma rays as it absorbs thermal neutrons. Results in the TFTR Test Cell were much in agreement with theoretical predictions and experimental measurements using activation foils.

Results from the EML measurements at the TFTR have been presented at several meetings, including the 36th Annual Meeting of the American Physical Society, Division of Plasma Physics, Minneapolis, November 1994.

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2.3 NEUTRON FIELD MEASUREMENTS AT THE U.S. ARMY PULSE RADIATION FACILITY

Paul Goldhagen, Ferenc Hajnal, Peter Shebell, Gladys Klemic and Nestor Azziz

As part of the reevaluation of the Hiroshima-Nagasaki atomic bomb survivor dosimetry, EML scientists participated in field experiments sponsored by the Defense Nuclear Agency at the Army Pulse Radiation Facility (APRF) of the APG in May 1992 and June 1993. They were asked to perform neutron field characterizations at various distances from the bare critical assembly (small unshielded fission reactor), which would provide benchmark data on neutron fluences and energy spectra to test long-range neutron transport calculations using codes similar to those applied to the Japanese A-bomb survivor dosimetry. At distances over 1 km from the epicenter of the Hiroshima detonation, where almost all the survivors were, the most recent transport calculations still predict a far lower fluence of thermal-energy neutrons than that determined from activation measurements (Kaul, 1993; Straume et al., 1992). Since most of the quantitative knowledge we have about the long-term effects of radiation on people comes from studies of the Japanese atomic bomb survivors, the resolution of this discrepancy is one of the most important problems in radiation dosimetry.

For both the 1992 and 1993 measurements, neutron fluences and energy spectra were obtained using the EML multisphere neutron spectrometer with 12 detectors, each of which contains a BF_3-filled pulse ionization chamber. The 1992 measurements were made at distances of 715, 1084, and 1588 m from the critical assembly right after several days of heavy rain. In 1993, the distances were 300, 1588, and 1986 m, and the ground was relatively dry. Gamma-ray energy spectrum and dose-rate measurements were also taken in 1993, using an intrinsic germanium de-

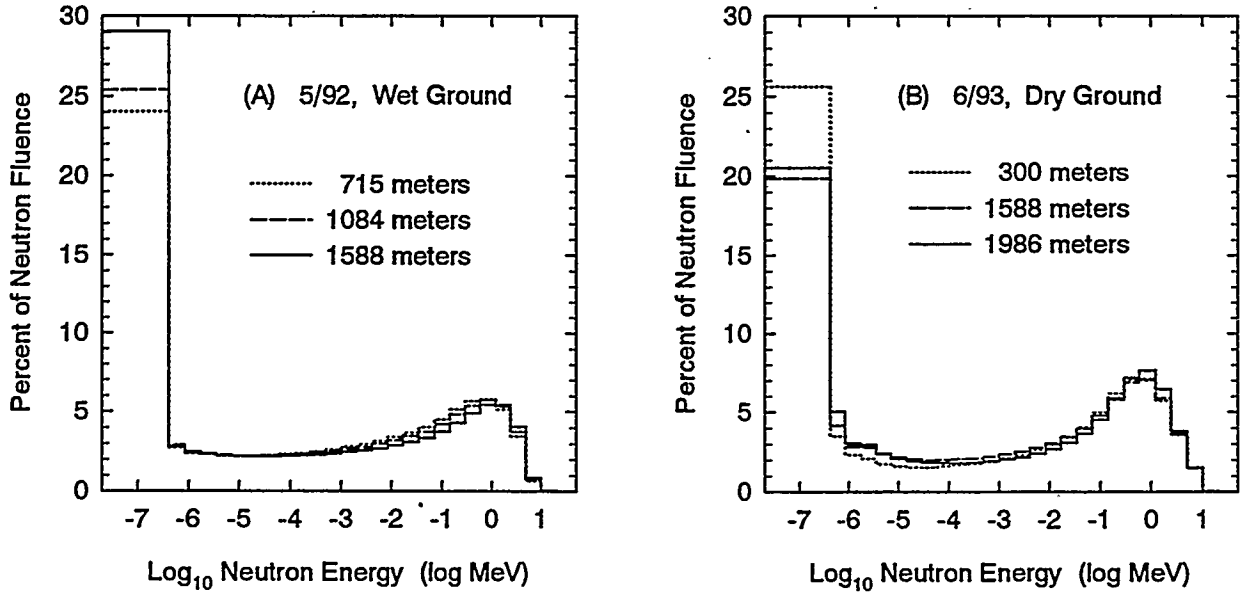

Figure 2.2 Relative neutron fluence as a function of energy measured by EML in in 1992, (a) and again in 1993, (b) at various distances from the APRF unshielded uranium critical assembly.

tector and pressurized ionization chambers. In addition, thermal neutron fluences were measured using a new Al_2O_3 thermoluminescent dosimetry system as a test of the system (Azziz et al., 1994).

Preliminary neutron fluence, kerma and dose equivalent spectra were determined this past year using our older set of energy response functions for the detectors. Relative fluence vs. energy spectra for the 1992 and 1993 measurements are shown in Figure 2.2 (A) and (B). These preliminary results show that for each year, the shape of the spectrum does not change much with distance, although there is a change from one year to the next. Presumably, the higher fraction of thermals in the 1992 measurements is due to the high ground water content at that time (11-38% by weight). In each year, the total dose equivalent per unit neutron fluence varies by only about 5% or less as a function of distance. Final APRF neutron energy spectra will be unfolded from our data when the results of the recent calculations for the energy response of each of the multisphere neutron spectrometer detectors (see Summary No. 2.4) are incorporated into our spectral unfolding code.

Preliminary results for the thermal neutron fluence, reported in this section last year, showed a falloff with distance closely matching that for the thermal neutrons at Hiroshima determined from activation measurements. This suggests that the energy spectra of the APRF fission neutrons and the long-range Hiroshima neutrons were similar, and hence that the discrepancy between calculations and measurements of thermal neutron activation at Hiroshima may involve some higher-energy fission-like neutrons emitted from the weapon rather than any problems with the long-range transport calculations. If the Hiroshima survivors were exposed to significant numbers of fast neutrons, and not just to gamma rays, estimates of the health risks from exposure to both neutron and gamma radiation would be affected.

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2.4 CALCULATION OF MULTISPHERE NEUTRON SPECTROMETER RESPONSE FUNCTIONS

Paul Goldhagen

EML has been measuring neutron energy spectra for over 20 years, most recently at the TFTR, the APRF and on a Canadian Forces aircraft in flight (see Summaries Nos. 2.2, 2.3 and 2.7). The primary instrument for these measurements is a multisphere neutron spectrometer, which is a series of spherical polyethylene moderators of different sizes surrounding counters sensitive to slow neutrons. The slow-neutron counters of the multisphere neutron spectrometers used for most of the measurements since 1985 are pulse ionization chambers filled with $^{10}BF_3$ gas. Determining a neutron energy spectrum from the number of counts in each of the detectors requires knowing each detector's efficiency as a function of energy, called its response function. Until recently, we have been using response functions based on calculations made by ORNL in 1971 (Maerker et al., 1971). Now we have performed new calculations of the response functions of our multisphere neutron spectrometer to improve the accuracy of the energy spectra we can obtain.

In the new calculations, the latest available neutron interaction cross sections (ENDF/B-V) were used, along with the Monte Carlo particle transport code MCNP (Briesmeister, 1993), which allows detailed modeling of the actual detector geometry.

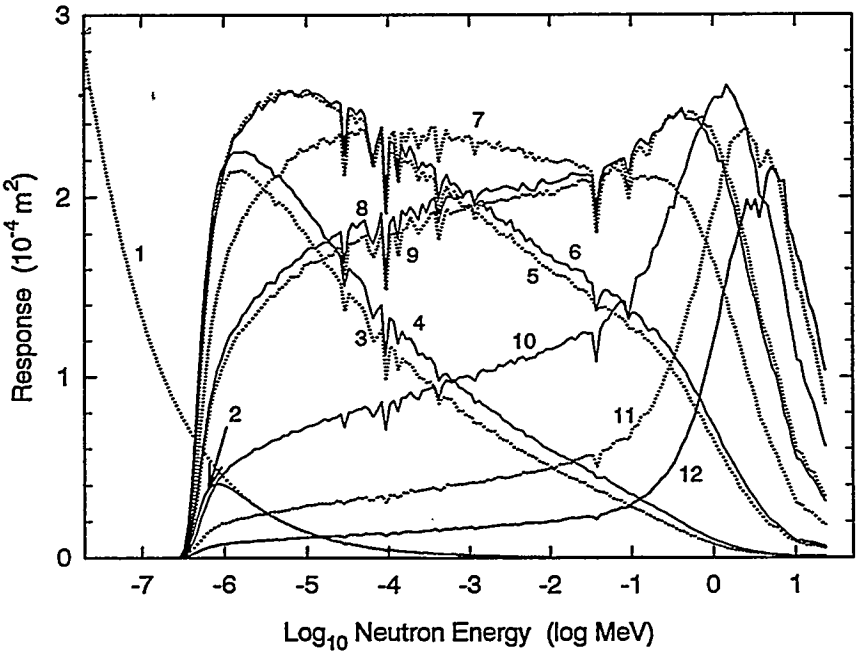

Figure. 2.3 Calculated response functions for the twelve detectors of the EML multisphere spectrometer with $^{10}BF_3$ counters. The vertical axis of effective area, 10^{-4} m^2, are equivalent to counts cm^2 neutron^{-1}.

Results of the new calculations for all 12 detectors of the Multisphere neutron spectrometer with $^{10}BF_3$ counters are shown in Figure 2.3. The calculations were done with a continuous distribution of incident neutron energy, with the results grouped into 20 bins per energy decade.

The results are properly viewed as histograms, but we have drawn them as continuous lines for visual clarity. The sharp dips come from absorption resonances in the cadmium and aluminum shells surrounding the detectors. (The detectors are surrounded by cadmium to reduce their response to incident thermal-energy neutrons.) The new response functions differ from the old ones by up to 25%, mostly for detectors 3 through 6, which have the smallest moderators.

When the new response functions for our existing MSNS are grouped into wider energy bins and incorporated into our spectral unfolding code, we will use them to unfold neutron energy spectra from our recent experimental data to see if the spectra and consequent dose equivalents are significantly different from the previous results. We will also calculate the response of our new multisphere neutron spectrometer with ^{3}He proportional counters (see Summary No. 2.5), and, in collaboration with LANL, extend the calculations to higher energies using the Los Alamos High Energy Transport Code (LAHET).

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2.5 New Multisphere Neutron Spectrometer

Paul Goldhagen and
William Van Steveninck

To perform cosmic-ray measurements in aircraft, a new multisphere neutron spectrometer has been built which is more portable, rugged, has higher neutron sensitivity, and a wider energy response than our earlier spectrometers. These characteristics have been achieved primarily by using high-pressure (4 atm) ^{3}He proportional counters as the slow-neutron sensors instead of the low-pressure (0.5 atm) $^{10}BF_3$ PICs used in the previous spectrometer. Both types of counters have 5-cm diameter spherical sensitive volumes. The ^{3}He counters have about 3 to 5 times the neutron sensitivity of the $^{10}BF_3$ counters, operate with a gas gain of 30 at 980 V, and have excellent discrimination between neutron and gamma ray/electron events. The high gas gain allows the detectors to tolerate 10 times more vibration than the $^{10}BF_3$ counters without producing spurious counts, while the relatively low operating voltage avoids electrical discharges in connectors, etc., at the low air pressures aboard aircraft. Portability, insensitivity to vibration and quick set-up time are enhanced by operating the detectors and their preamplifiers cushioned with plastic foam inside lightweight aluminum cases. A photograph of three of the detectors in their case is shown in Figure 2.4. Calculations of the response functions of the detectors will include the effects of the cushioning, cases and other detectors on the neutron field.

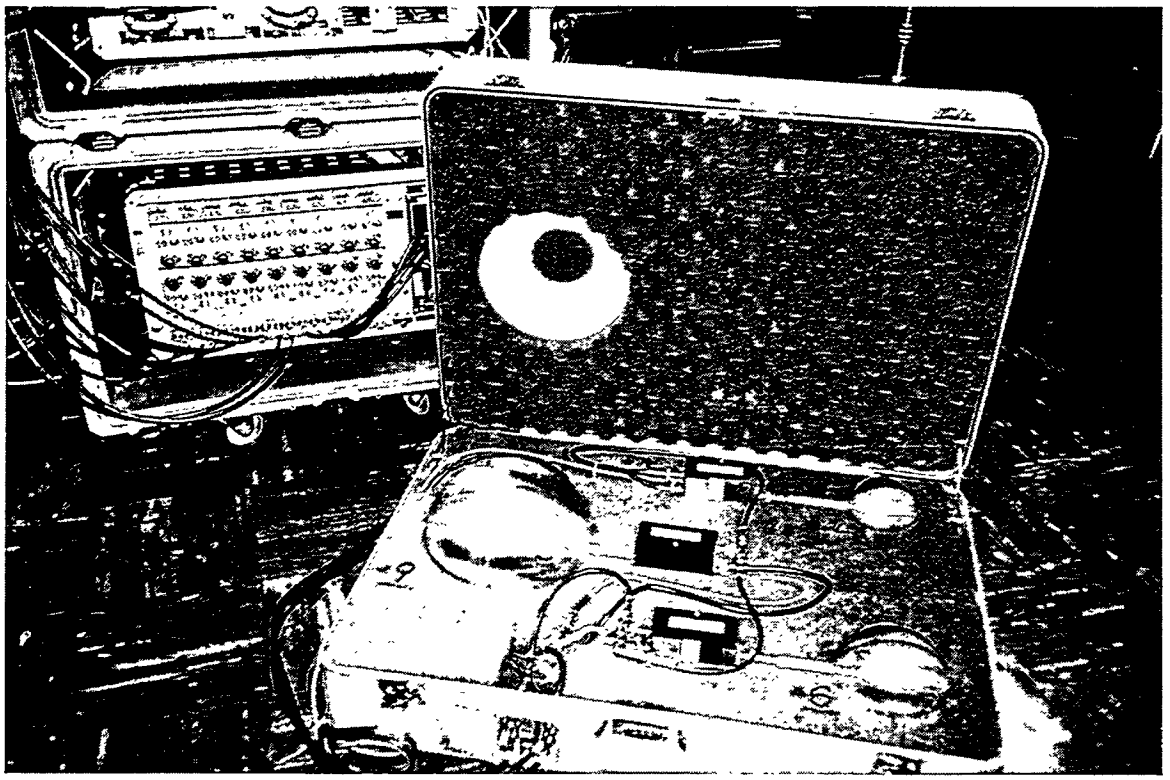

Figure 2.4 Photograph of the detectors of the new multisphere neutron spectrometer in their aluminum case in front of the EML amplifiers for the whole MSNS.

The new ^{3}He-counter spectrometer has 13 different detectors with a different array of spherical polyethylene moderator sizes than the $^{10}BF_3$- counter spectrometer. After the bare and cadmium-covered counters for detecting thermal and epithermal neutrons, the smallest new moderator is a shell that is only 0.75-cm thick. The moderator sizes increase so as to produce energy responses that increase in fairly uniform logarithmic steps. The moderator of the 11th detector is the same diameter (30 cm) as the largest moderator of the $^{10}BF_3$- counter spectrometer, and the moderator of the 12th detector is about 39 cm in diameter. The 13th detector has a polyethylene moderator 30 cm in diameter, but within the moderator is a 25-kg lead shell that boosts response to high-energy neutrons, which can produce several additional neutrons when they interact with lead nuclei.

EML's Instrumentation Division has designed and assembled amplifiers for the new multisphere neutron detector which use one fourth the electric power and take up half the space of standard commercial amplifiers. The EML amplifiers have a 6 μs shaping time to match the slow pulse-

formation time that is a trade-off consequence of the desirable characteristics of the ^{3}He proportional counters.

The new MSNS has been tested successfully during the first of several planned measurements of cosmic-ray neutrons aboard a Canadian Forces Boeing 707 aircraft (see Summary 2.7). Because of its portability, ruggedness, sensitivity and wide energy response, the new spectrometer will also be ideal for future low-level neutron measurements at accelerators and elsewhere.

2.6 RADIATION SAFETY FOR FUTURE HIGH-ALTITUDE COMMERCIAL AIRCRAFT

Ferenc Hajnal, Paul Goldhagen and Marcel Reginatto

Research in support of an economically competitive and environmentally safe high-speed civil transport (HSCT) is a high priority within NASA aeronautical programs. Designs are being studied for commercial aircraft that would cruise at altitudes of about 21 km (70,000 feet) -- near the Pfotzer maximum of cosmic ray particle flux in the atmosphere. Crews working on present-day subsonic aircraft flying at lower altitudes already receive one of the highest average dose equivalents of any occupationally exposed group in the U.S. Since the structure and operational characteristics of an aircraft affect the radiation environment of those within, the radiation safety aspects of a future high-altitude aircraft should be addressed early in its design phase.

In collaboration with the NASA Langley Research Center, we have examined the radiation safety aspects of the HSCT, including the traffic conditions envisioned for the aircraft, the radiation environment as it is now known, and the relevant health issues. Although the atmospheric radiation environment is better understood than when the commercial supersonic transport was proposed in the 1960s, present knowledge of the components of the radiation field which are biologically most important rests on theoretical predictions which have not been verified experimentally. There are large uncertainties (plus or minus a factor of 2) in our knowledge of the physical fields for high-energy neutrons and multi-charged ions, which need to be reduced. Furthermore, there is relatively little biological data on the effects of exposures to these components on which to base risk estimates. Improved methods for estimating risks in prenatal exposure need to be developed. A firm basis for solar flare monitoring and, if possible, forecasting needs to be developed, with means of exposure abatement. Results of this study will appear in a NASA Technical Paper (Wilson et al., 1995).

A program of high-altitude measurements is being developed to reduce the uncertainties in our knowledge of the neutrons and multiply-charged ions, and to provide benchmarks for calculations of the atmospheric radiation field. The instrumentation will include modified versions of the apparatus used for the measurements aboard Canadian Forces aircraft at altitudes typical of present-day air travel (see Summary 2.7).

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2.7 MEASUREMENT OF COSMIC RADIATION ABOARD A CANADIAN FORCES AIRCRAFT IN FLIGHT

Paul Goldhagen, Ferenc Hajnal, William Van Steveninck, Alfred Cavallo and Peter Shebell

EML has been invited to join the Royal Military College (RMC) of Canada and the Defense Research Establishment, Ottawa (DREO) in a collaboration to perform cosmic-ray measurements aboard dedicated flights of Canadian Forces CC-137 (Boeing 707) aircraft. The purpose is to characterize the cosmic radiation field in aircraft at altitudes typical of military transport operations and commercial air travel. EML was tasked with measuring the energy spectrum of the neutrons: the component of the atmospheric radiation field that contributes the greatest uncertainty to the dose equivalent and to estimates of health risk.

DREO and RMC are flying a variety of radiation detectors. EML is flying our new ^{3}He-counter multisphere neutron spectrometer (see

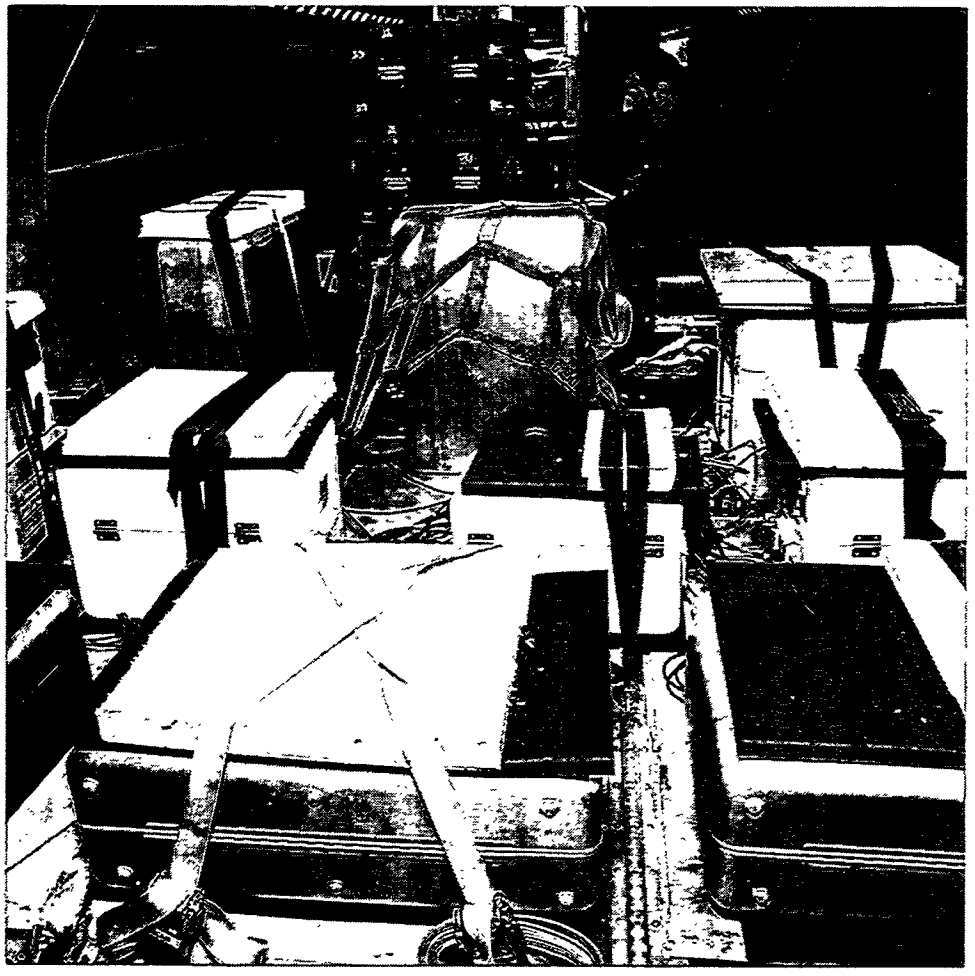

Figure 2.5. Measurements in progress aboard a Canadian Forces CC-137 (Boeing 707) 37,000 feet above Canada on 12/16/94. The white cases each hold one of the four largest detectors of the MSNS; the two aluminum suitcases standing on end hold the scintillators.

Summary No. 2.5), a PIC to measure total ionization for flight-to-flight normalization, and plastic, sodium iodide, and bismuth germanate scintillators with anti-coincidence shells. The anti-coincidence shells discriminate between the charged and neutral components of the incident radiation, while the different materials and densities of the inner scintillators should enable us to separate the nucleons from the leptons and gamma rays. Such measurements, useful in their own right, are needed to correct the multisphere neutron spectrometer results for response to high-energy protons. A photograph of some of the EML apparatus aboard the aircraft is shown in Figure 2.5.

A successful test flight was made over Canada in December, 1994. The flight was at a constant pressure-altitude of 11.3 km (37,000 feet) for 4 h, and at a constant geomagnetic latitude (~75 N) for most of that time to sample a nearly constant radiation field. The ionization dose rate in air measured by the PIC was approximately 2.4 mGy h^{-1}, and it varied by < 1% during the constant-altitude portion of the flight. The total neutron fluence was about 250 times its value on the ground, and neutron spectral data with thousands of counts in each detector were recorded every 5 min. Two round-trip transatlantic flights are planned in 1995-96. These flights should provide high-quality neutron energy spectra extending up to 1 GeV and a great deal of other data at altitudes typical of present-day air travel.

2.8 CALCULATION OF DOSE, DOSE EQUIVALENT, AND RELATIVE BIOLOGICAL EFFECTIVENESS FOR HIGH CHARGE AND ENERGY ION BEAMS

F. Hajnal and M. Reginatto

There are several practical applications in which exposures with high charge and energy (HZE) ions are of concern. Among these are the natural cosmic-ray environment in space, consisting of relativistic nuclei of all elements, for which HZE ions contribute 80% of the free space dose equivalent. Even for modest depths of shield penetration (30 g cm^{-2}) the dose equivalent of the HZE ions and the secondary particles they generate is on the order of 45% of the total, and therefore of considerable importance in assessing the radiation protection needs of future high-speed high-altitude aircraft. Computational models are also required for treatment planning in cancer therapy with HZE accelerator beams, in which cell killing is the critical parameter. With the further development of HZE accelerators, there is a growing concern among policy setting bodies on evaluation of exposures for the scattering of HZE particles from the beam line.

The biological response of living tissues depends in part on the temporal and spatial fluctuations of the energy deposits within the

tissue system. Such fluctuations depend not only on the specific radiation environment to which the person is exposed, but also on how that environment is modified by interaction with the human body in reaching specific tissues. Even if the radiation environment to which the individual is exposed is known precisely, the energy deposits within specific tissues deep in the body are largely known through theoretical estimates and are therefore limited by the uncertainty in the calculational model. Clearly, an accurate conversion of the radiation environment to estimates of the exposure fields at specific tissue sites is an important issue in HZE radiation protection problems

In a collaboration with NASA Langley Research Center, analytical methods developed at NASA Langley (Wilson et al., 1990) were applied to evaluate dosimetric quantities as a function of penetration depth in tissue equivalent materials to estimate specific organ exposures. The Green's function for the transport of ions of high charge and energy was used to evaluate dose, dose equivalent, and relative biological effectiveness for C3H10T1/2 mouse embryo cell survival and neoplastic transformation as a function of depth in soft tissue. Many of the qualitative features of the relative biological effectiveness were represented by the average quality factor, but important track structure dependent differences were observed. Differences in the values of the relative biological effectiveness for cell survival and for neoplastic transformation (which are the result of specific track structure parameters associated with survival and transformation) were consistent with the data of Yang et al. (Wilson et al., 1993). A paper on this subject was submitted to the Health Physics journal in 1994 (Wilson et al., in press).

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2.9 MEASUREMENT OF THE NEUTRON RESPONSE OF VARIOUS TLDs

Gladys Klemic and Nestor Azziz

The recently developed phosphors Al_2O_3:C and ^{7}LiF:Mg,Cu,P are attractive for use in personnel and environmental dosimetry because of their very high photon sensitivity. For potential application in mixed neutron-gamma fields, we investigated the relative neutron response of these thermoluminescence dosimeters (TLDs) compared to that of the widely used ^{7}LiF:Mg,Ti. Monoenergetic neutrons of energies from 0.33 to 14 MeV produced by a Van de Graaff accelerator at the Radiological Research Accelerator Facility (RARAF) of Columbia University were used to irradiate pairs of TLDs (see Figure 2.6).

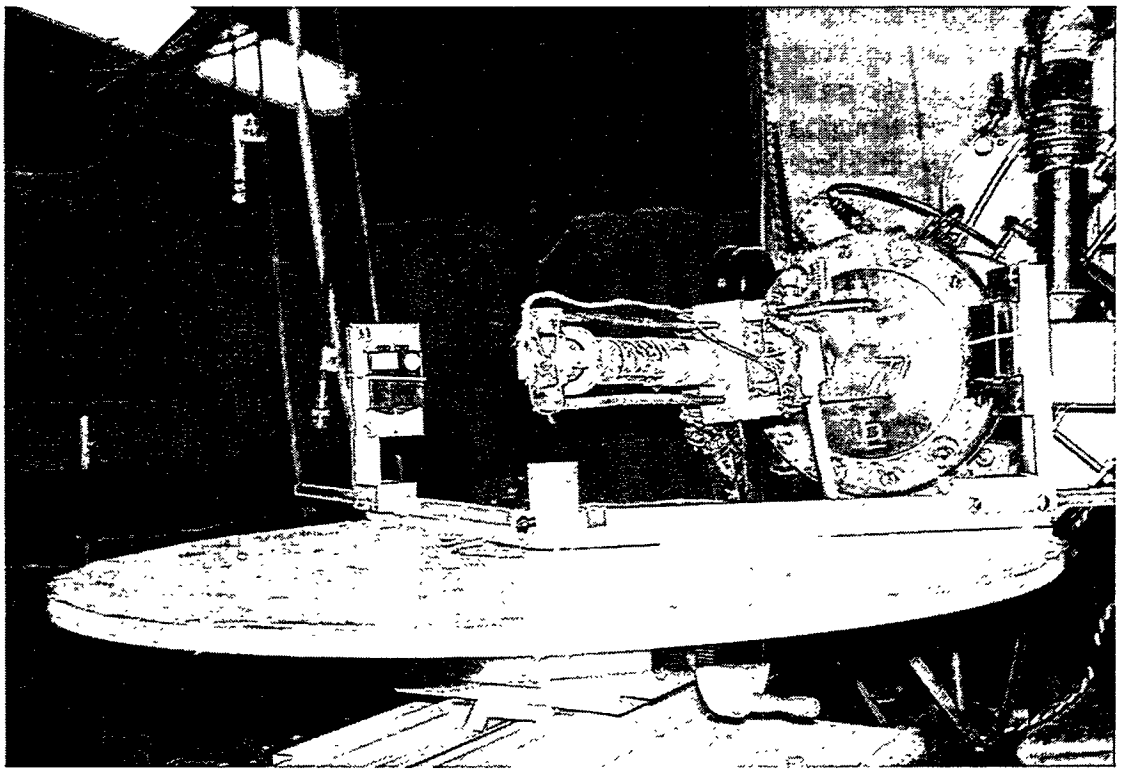

Figure 2.6 Experimental platform for irradiation of samples with essentially monoenergetic neutrons at RARAF.

To isolate the TLD's response to neutrons, it is necessary to fully account for all gamma dose resulting from photon production in the RARAF target as well as that due to natural background radiation. Control TLDs were used to measure the exposure received in transit and storage, and other controls were used to calibrate the response to gamma radiation using a ^{137}Cs source. A tissue equivalent ionization chamber was used to measure the total absorbed dose (mrad) received in the RARAF beam, while a compensated Geiger-Muller chamber separately determined the gamma component. Gold activation foil pairs (bare and cadmium covered) were used to check for any thermal neutrons that could result from room scatter.

TLDs were read out using a manually operated, linearly heated planchet reader, and the activation foils were counted using a high-purity germanium detector. Preliminary analyses indicate that the relative neutron sensitivity (neutron: gamma) of ^{7}LiF:Mg,Cu,P is an order of magnitude less than that of ^{7}LiF:Mg,Ti , while that of Al_2O_3:C may be much lower. Analysis of aluminum oxide neutron sensitivity is complicated because the neutron response is near the lower limit of detection of our system. The initial results indicate that these new materials could be useful for discriminating neutron and gamma components of dose in mixed radiation fields. This work will be presented at the upcoming 11th Solid State Dosimetry Conference.

2.10 PORTABLE TLD READER

Nestor Azziz, Michael Polito,
Frederic Guggenheim, and
Vincent C. Negro

There has been a significant increase in the use of TLDs by EML as gamma and neutron detectors in a variety of applications (see Summaries 1.10, 2.1, 2.2, and 2.9) to improve the ease and accuracy of TLD measurements made in the field, a portable TLD reader is being developed. As described in last year's report, this reader will be especially advantageous for low-level measurements at remote sites, since its use avoids relatively large backgrounds otherwise accumulated during transport to and from the site.

The portable reader uses a nickel-plated soldering gun tip for its heating element, a miniature photomultiplier tube with integral high-voltage supply as its sensor, and a model 5F-LCD Tattletale microcomputer for controlling operation and recording the data. Initial tests of the new reader have been promising, and several defects were detected and corrected. Problems maintaining a linear temperature ramp were reduced by using a more complex heater power supply, and the deviation from linearity is now less than 1%. Above 300° C, the heating tip glowed, interfering with the light signal from the TLD. This problem was reduced by a smooth plating of the tip and will be further reduced by a stainless steel shield. Glow curves recorded using the new reader are acceptable, but the results are not yet repeatable. Further testing will continue with emphasis on improving the software of the Tattletale microcomputer that drives the reader.

3

Environmental Radon, Thoron, and Related Aerosols

3.1 OVERVIEW

Earl O. Knutson

The EML Radon Research Program for 1994 consisted of four major research areas as follows:

Quality Assurance:

EML serves as a Quality Assurance /Quality Control (QA/QC) center for the OHER Radon Research Program. EML is also one of four regional reference laboratories in the International Intercalibration and Intercomparison Program for Radon, Thoron, and Daughters Measuring Equipment (IIIP) of the International Atomic Energy Agency (IAEA), serving as a regional intercomparison center for North American radon, thoron and progeny measurements. The IIIP is now part of the IAEA International Radon Metrology Program (IRMP).

Instrument Development:

Radon measurement technology at EML currently focuses on the development of small and portable devices for indoor radon and thoron concentration measurements, and on instrumentation for measuring very low levels of radon gas.

Aerosol Physics:

The aim of the aerosol physics project is to identify and quantify the physics and chemistry processes that determine the particle size spectra of indoor radon and thoron progeny aerosols. This information is valuable to those who seek ways of reducing the radiological dose to the human respiratory tract. Processes that produce ultrafine (<10 nm) or coarse (>1μm) particles are especially important since these sized particles have high dose conversion factors. Processes that alter the size of particles over time, such as coagulation, are also important.

Exposure Studies:

The differences in the exposure between modern homes and the old uranium mines, especially those factors most significant in determining the impact on human health from radon exposure has been the focus of this research over the past year.

Quality Assurance Activities

3.2 NATIONAL RADON GAS INTERCOMPARISONS

Isabel M. Fisenne, Andreas C. George and Pamela Perry

QA/QC are the backbone of many commercial and research programs and processes.

The quality of the radon measurements in the U.S. have been tested under controlled conditions in semi-annual radon gas intercomparison exercises sponsored by EML since 1981. OHER mandates the participation of its radon contractors in appropriate EML radon and progeny intercomparisons.

EML's reputation as a premier laboratory in QA/QC methodology and environmental radon, progeny and aerosol measurements has attracted over 325 participants from a pool of 76 groups, including 11 foreign institutions to participate in our exercises over the past 13 years.

The 25th radon intercomparison exercise (April 1994) drew 27 participants. The mean radon concentration results obtained at EML for 22 samplers measured in the EML pulse ionization chamber (PIC) was 570 ± 12 Bq m^{-3}, and 566 ± 5 Bq m^{-3} for four scintillation cells. Twenty-one of the 27 participants (78%) reported mean radon concentration values within ±10% of the PIC value. All the reported means were within ±25% of the PIC value.

Twenty-six groups participated in the 26th radon intercomparison exercise (October 1994). The radon concentration determined from 18 samplers measured in the PIC was 945 ± 15 Bq m^{-3} and 935 ± 29 Bq m^{-3} for four scintillation cell measurements. Twenty-four groups (79%) were within ±10% of the EML PIC value, and 96% of the groups were within ±25% of this value.

The 25th and 26th radon intercomparison exercises followed previous patterns in which the majority of the groups were within ±10% of the EML value. There was still one outlier, demonstrating the need for some form of independent QA/QC evaluation.

We were pleased to have the participation of the Czech Reference Laboratory for Radon and Daughters, Milin, Czech Republic; and the National Atomic Energy Commission, Buenos Aires, Argentina; as well as our Canadian and Swiss colleagues. This reflects the continued support of the scientific community for the QA/QC programs performed at EML.

The 1994 radon intercomparisons, while on the whole are within acceptable limits, still point out the need to continue the program and to coordinate our QA/QC efforts with our European colleagues. This is especially true since validation information must be collected contemporaneously with field measurements. EML's efforts in QA/QC provided independent, documented evidence of the validity of the U.S. database for radon and progeny measurements. This documentation is necessary so that at sometime in the future this database can be used with confidence.

3.3 INTERCOMPARISON OF INSTRUMENTS AND METHODS FOR MEASURING RADON AND RADON PROGENY IN INDOOR AIR

Andreas C. George, Keng Wu Tu and Earl O. Knutson

An intercomparison exercise for radon and radon progeny instruments and methods was held at EML from April 22-May 2, 1994. The exercise, part of the International Radon Metrology Program (IRMP), was conducted in the new EML radon, thoron and progeny exposure facility (Fisenne and George, in press) in which conditions of exposure are very well controlled.

The intercompared instruments included: 1) pulse ionization chambers, 2) electret ionization chambers, 3) scintillation detectors, 4) alpha particle spectrometers with silicon diodes, surface barrier or diffused junction detectors, 5) registration of nuclear tracks in solid-state materials, 6) activated carbon collectors counted by gamma-ray spectrometry or by alpha and beta liquid scintillation counting.

For radon gas, there were 23 participants including private firms, government laboratories, and universities. Altogether, 165 passive integrating devices were tested, including activated carbon collectors, nuclear alpha track detectors, electret ionization chambers, and 11 continuous radon monitors. For radon progeny, five portable instruments were intercompared. Also, the five groups that participated in person collected a total of 40 grab samples for radon progeny.

The results indicate that more than 80% of the measurements for radon gas were within ± 10% of the EML reference value. The majority of the instruments that measure individual radon progeny and potential alpha energy concentration (PAEC) gave results that were in good agreement with the EML reference value. Radon progeny measurements made with continuous and integrating instruments were satisfactory, but there is room for improvement.

The results of the intercomparison were reported in a Radon Workshop on Indoor Air "An Integrated Approach" held in the Gold Coast of Australia, November 27 to December 1, 1994. The paper will be published in the Proceedings of the Workshop (George et al., in press,*a*). A second and more detailed report will be published as an EML report (George et al., in press,*b*).

References

George, A. C., K. W. Tu and E. O. Knutson "Third Indoor Radon and Radon Progeny Measurements Intercomparison of Instruments and Methods" Proceedings of the International Workshop on Indoor Air - An Integrated Approach, in press,*a*

George, A. C., K. W. Tu and E. O. Knutson "Intercomparison of Active and Passive Instruments for Radon and Radon Progeny in North America" USDOE Report EML-566, in press, *b*

Fisenne, I. M. and A. C. George "Radon, Thoron, and Progeny Exposure Facilities" EML Procedures Manual, HASL-300, 28th Edition, Section 6.2.3., in press

3.4 INTERNATIONAL RADON METROLOGY PROGRAM (IRMP)

Andreas C. George

The aim of the IRMP is to provide the international scientific community with a network of reference and support services necessary to maintain high standards in the metrology of radon, thoron and their decay products. An example is the involvement of the IRMP in the IAEA/CEC Coordinated Research Program on Radon in the Human Environment, which involves 55 nations. In addition, the IRMP provides technical training and advice on the use of the state-of-the-art instrumentation for radon surveys and radon scientific research. The IRMP intercomparisons will ensure consistency in radon measurements on a global scale. Member laboratories of the IRMP are the four laboratories of the IIIP, two laboratories from the U.S. Environmental Protection Agency (EPA), and seven regional coordinating laboratories. EML is a charter member of the IIIP, which was established in 1983.

Andreas C. George represented the U.S. at an IRMP meeting of the Coordinated Research Program on "Radon in the Human Environment" sponsored by the IAEA in Vienna, December 12-16, 1994. At the IRMP meeting, the results of 51 research projects on national radon surveys and measuring techniques were presented by representatives from 26 countries. These findings form a firm and reliable background for future IAEA activities in this important fields of chronic exposure to natural radiation in different environments.

Andreas C. George wrote a section of an IAEA document on the design criteria and operational characteristics of radon, thoron and progeny calibration facilities for the IRMP. He also is in the process of collecting and documenting the design and operational characteristics of over 35 radon test facilities worldwide.

3.5 INTERCOMPARISON OF IMPACTORS AND A SCANNING MOBILITY PARTICLE SIZER FOR MEASURING RADON PROGENY PARTICLE SIZE

Earl O. Knutson and Keng Wu Tu

During the week of January 4-6, 1994, EML was joined by Professor Philip Hopke of Clarkson University in an intercomparison of radon progeny particle size measurements made with cascade impactors and a scanning mobility particle sizer (SMPS). Both groups have learned in recent years that modern, wide-size-range impactors have important advantages over diffusion batteries in sampling indoor radon progeny aerosols. This intercomparison marked the first use of EML's new radon, thoron and progeny exposure facility and its associated aerosol generation and characterization instruments.

EML used a 30 L min^{-1} MOUDI impactor, along with a separate reference filter. A graded screen array (GSA) was also used to better sample the ultrafine particles in the aerosol. Twelve tests were carried out using Carnauba wax aerosols of different particle sizes. The SMPS (see Summary No. 3.8) was used as the reference method for measuring the particle size. Table 3.1 shows the generally good agreement between the SMPS and MOUDI results.

Prof. Hopke used a 5.0 L min^{-1} Berner impactor. Of the two impactors, the one with a backup filter (the MOUDI) appeared to generate

TABLE 3.1

COMPARISON OF RESULTS FROM THE SMPS AND MOUDI

	SMPS		MOUDI	
Test ID	GMD* (nm)	GSD**	GMD*(nm)	GSD**
94010410.18	218	1.19	181	1.25
94010511.24	228	1.20	183	1.27
94010414.35	270	1.34	208	1.38
94010416.24	352	1.16	316	1.25
94010509.44	87	1.25	85	2.01
94010511.12	82	1.28	85	1.97
94010512.19	76	1.23	110	1.96
94010514.26	106	1.21	99	1.80
94010515.39	108	1.23	104	1.91
94010516.48	105	1.23	97	1.82
94010610.11	523	1.09	396	2.13
94010611.16	506	1.13	415	1.29

* GMD: geometric mean diameter.
** GSD: geometric standard deviation.

more stable, reproducible results. The smoother and flatter impaction surfaces of the MOUDI may also have played a part. Only two of the 12 tests showed close agreement between the two impactors.

3.6 COORDINATED RESEARCH AND CONSULTATIONS FOR THE DEVELOPMENT AND EVALUATION OF INSTRUMENTS AND METHODS

Andreas C. George, Earl O. Knutson and Keng Wu Tu

EML continues to share its knowledge and its radon, thoron and progeny test facilities with investigators that develop, upgrade, or refine instruments and methods for the assessment of the radiation dose to the general public. QA radon exposures were conducted with scintillation cells for the University of Florida, Bowser and Morner, Inc., the Pennsylvania Department of Environmental Resources, the New Jersey Department of Environmental Protection, and the New York State Department of Health. Prototype activated carbon collectors for radon were evaluated for the Pakistani Institute of Science and Technology, the Hebrew University of Jerusalem, the Jicarilla Apache Tribe, Thermoanalytical, Inc., and the states of New Hampshire, New Jersey, New York and Pennsylvania. New design electret ionization chambers for short-term and long-term exposures were tested and evaluated for the Radon Corporation of America and Rad Elec. Nuclear track detectors of different configurations were

evaluated for Altrac of Germany, and Landauer, Inc., Continuous radon monitors were tested for Bowser and Morner and for EPA, and continuous working level monitors were tested for AT&T and Radon QC.

Andreas C. George, as the technical program co-chair, played a leading role in the organization of the 1994 International Radon Symposium sponsored by the American Association of Radon Scientists and Technologists. He participated in several meetings to draft a part of ANSI N13.34 on the Performance Specifications for Methods and Instruments for the Measurement of Indoor Radon, and he wrote the section on the performance specifications for activated carbon collectors. Andreas C. George also participated in several meetings of the Radiological Control Coordinating Committee Radon Subcommittee to draft a Pre-decisional Report on Occupational Exposure to Radon and Thoron in DOE, sponsored by DOE EH-41, EM-25 and AL HPD.

Andreas C. George, in cooperation with Westinghouse Savannah River Site, tested the feasibility of using a sintered stainless steel filter to monitor airborne radioactive contamination at the Savannah River Operations. The sintered filter is only 15%-30% efficient for radon progeny and very efficient for fission products, which is a desirable feature in eliminating false alarms at such operations. More work is needed to determine the loading characteristics of the sintered filters.

Andreas C. George, as a member of an advisory committee, served as a principal consultant to the American Water Works Association in a project to assess radon progeny exposure while showering with radon bearing water.

At the invitation of the Health Physics Society, Andreas C. George prepared a paper entitled "State-of-the-Art Instruments for Measuring Radon /Thoron and Their Progeny in Dwellings" for presentation at the Health Physics Society 39th Annual Meeting in San Francisco, CA, and also for publication in the Journal.

Instrument Development

3.7 ACCEPTANCE TESTS OF THE EML RADON CHAMBER

Earl O. Knutson

As described in last year's annual report, a new radon, thoron and progeny exposure facility was installed at EML. The design criteria for the new chamber were: at least 25 m^3 useable volume; temperature controllable from 5° C to 40° C (set point held to ±1° C); dew point controllable from -10° C to 1° C below the temperature (set point held to ±2 percent relative humidity, RH); temperature/dew point changes complete in < 4 h; air leakage < 28 L min^{-1} at 5 Pa overpressure; aerosol loss rate minimization (goal: < 20% h^{-1}); unwanted aerosol minimization (goal: < 1000 cm^{-3}); organic vapors minimization.

During 1994, extensive testing was done to determine if the chamber can meet specifications. The main results of these tests were:

- Background aerosol: <200 cm^{-3}, as measured by an ultrafine condensation particle counter (lower concentrations, down to 1 particle cm^{-3}, can be achieved by increasing the pressurization air stream to 120 L m^{-1} of highly filtered air).
- Air leakage: in the range 26 to 39 L min^{-1}.
- Aerosol loss rate: approximately 0.6 h^{-1}, dropping to 0.14 h^{-1} when the internal blower is turned off .
- Temperature and humidity control: acceptance testing is not yet complete.

Concurrent with these tests, the chamber was in use for most of 1994. Examples of the chambers's use can be found elsewhere in this report (see Summaries Nos. 3.3, 3.5, 3.8, 3.10, 3.11, 3.20).

3.8 AEROSOL GENERATION/ MEASUREMENT FACILITY FOR EML's RADON CHAMBER

Keng-Wu Tu

Two condensation monodisperse aerosol generators (Model 3472 and 3470, TSI, Inc., St. Paul, MN) were purchased, installed, and tested this year in EML's new radon, thoron and progeny exposure facility. The objective was to produce Carnauba wax particles with narrow size distributions for use as test aerosols. The ability to generate well-defined test aerosols is important for conducting radon and related aerosol research. These two aerosol generators are each able to provide near monodisperse condensation aerosols at the sizes ranging from 60 nm to 1100 nm, with number concentrations up to 50000 cm^{-3} in the new radon chamber. The two generators can also be operated simultaneously to produce bimodal aerosols. The Model 3472 generator was designed for high flow rate, up to 15 L min^{-1}, while the Model 3470 is only up to 4 L min^{-1}. The size and monodispersity of the particles were checked with TSI's electrostatic classifier (Model 3071A). Listed in Table 3.2. are the particle sizes (geometric mean diameter, GMD), condensation nuclei (CN) concentrations in the chamber, and the corresponding generation conditions, including oven temperature (T_{OV} °C), reheating temperature (T_{reh} °C), and N_2 flow rate (Q_N). The geometric standard deviations (GSD)

TABLE 3.2

CARNAUBA WAX PARTICLE SIZES VERSUS GENERATION CONDITIONS FOR TWO CONDENSATION MONODISPERSE AEROSOL GENERATORS

	MODEL 3470				MODEL 3472			
Particle Diameter (nm)	T_{ov} (°C)	T_{reh} (°C)	Q_n (L min^{-1})	CN (cm^{-3})	T_{ov} (°C)	T_{reh} (°C)	Q_n (L min^{-1})	CN (cm^{-3})
45	110	115	3.58	-	-	-	-	-
60-70	130	1135	3.5	10k	100	110	3.5	20k
90-100	140	145	3.5	15k	120	125	3.5	50k
150	170	175	3.5	25k	130	135	4.0	50k
300-350	190	200	3.5	40k	140	150	2.5	30k
500	210	215	3.5	30k	200	205	4.5	40k
11000	-	-	-	-	250	260	6.5	10k

were typically around 1.2-1.3 measured at the output of the generators. The GSD's became larger, ranging from 1.3 to 1.6, when the particles were measured in the chamber because of the background aerosols. The GSDs for the particles in the chamber therefore depend on the concentration of background aerosols. Figure 3.1 shows four representative samples, A and C are measured at the output of the generators, while B and D are measured in the chamber. When the background aerosol concentration is low, a few hundreds of particles per cm^3, both curves of a set are quite similar, and the GSDs of the two curves are very close, 1.25, such as curves A and C. However, when the background aerosol concentration is high, several thousands of particles cm^{-3}, the GSD became larger, such as in curve B.

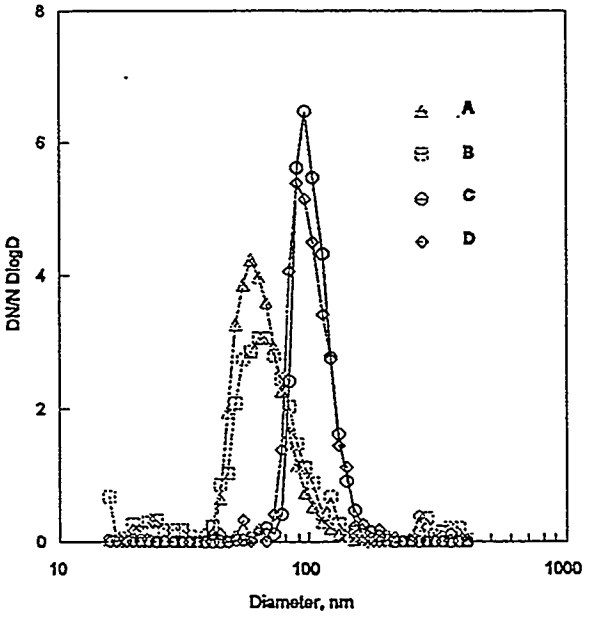

Figure 3.1 Particle size distribution of Carnauba wax aerosol.

3.9 BARREL RADOMETER

Vincent C. Negro, S. Frederick Guggenheim, Michael Polito and Joseph Ventre

At present EML has atmospheric radon monitors in use at Mauna Loa, Hawaii and Bermuda (see last year's report, p. 49). They are large two-filter instruments, 1 m in diameter and 2 m to 3.5 m long , having decay volumes of 500 L and 1000 L. Because of their size and bulk, they are difficult to transport and require 2 or 3 people to deploy.

A new "Barrel Radometer" instrument has been developed for measuring low-level radon concentrations. The Barrel Radometer fits in a 182-L drum, making it easy to transport and deploy. While the sensitive volume of this instrument is only 100 L, it uses counting-while-sampling as opposed to counting-after-sampling used in two filter instruments. This along with the low background of spectroscopy detection combine to yield a sensitivity equal to or better than that of a 500 L two-filter instrument.

The operating principles of the Barrel Radometer are the same as the Radometer II (see last year 's report, pp. 26-27). Charged radon progeny are attracted by an electric field to a solid-state detector where further alpha decay is registered using alpha spectroscopy. However, due to the larger volume, the high voltage used for the attraction field was increased over that of the Radometer from 3 kV to 30 kV. In addition, to retain the hemispheric geometry, the 100-L cylinder is split into two 50-L cylinders by a screen at the midpoint. Each of these 50-L sections has a separate detector, and electronics are used to combine the data. Because moisture neutralizes the charged progeny, a small air conditioner is built in to keep the dew point low.

High voltage tests have been successfully completed and most of the fabrication work has been done. The instrument should be ready for testing and calibration by late summer of 1995.

3.10 A PORTABLE, BATTERY-POWERED, CONTINUOUS AIRBORNE ^{222}Rn SAMPLER

Salvatore C. Scarpitta

A 27 ounce, battery-powered, sampling system, using a multiple port sampler (Polyport; Polito and Albert, 1994) was designed and constructed at EML. In this sampler, ^{222}Rn is collected in one of 12 6-cm long glass tubes containing 0.4 g of Carboxen, a hydrophobic beaded carbon molecular sieve (BCMS) material (see Figure 3.2). After sampling, adsorbed ^{222}Rn progeny, ^{214}Pb and ^{214}Bi are measured using either a gamma detecting NaI crystal or a liquid scintillation counter.

The sampler was tested to determine its effectiveness in collecting ^{222}Rn gas in humid air. Experiments were initially conducted in the EML radon chamber to determine the optimum sampling conditions. The variables tested were sample flow rate (SFR), temperature, sampling time and RH. The first three of the variables had a minimal effect on ^{222}Rn collection efficiency, but high RH reduced the ^{222}Rn adsorption by 70%. A single calibration curve was developed to account for the adverse effects of RH on ^{222}Rn adsorption over a 15 degree range in temperature (i.e., 20 to 35°C).

This calibration curve was then used in two field tests to determine the average hourly ^{222}Rn levels based on the amounts of both ^{222}Rn and water vapor adsorbed by the BCMS material. Results obtained (Table 3.3) with the Polyport were compared to those obtained with EML's Radometer II (see last year's report; Negro, 1990). With liquid scintillation counting, the minimum detectable ^{222}Rn concentration for both hourly samples and hourly measurements was about 6 Bq m^{-3} (0.22 pCi L^{-1}) at 50% RH, when a sample flow rate of 5 L h^{-1} was used. At 20% RH and room temperature, the BCMS material collected ^{222}Rn with 60% efficiency.

The EML Polyport system can be deployed in homes, atmospheric balloons or in remote areas where a power supply is not available and continuous hourly measurements are desired. The lower limits of detection for the gamma counter and the liquid scintillation counter were 0.4 Bq for a 0.5 h count time and 6 mBq for a 1 h count time, respectively. The results of this study will be submitted to the Health Physics Journal early next year.

TABLE 3.3

FIELD TEST AT EML BASEMENT (RH = 20%)

Tube ID	Polyport (Bq m^{-3})	Radometer II (Bq m^{-3})
A	30.0	33.6
B	30.4	25.9
C	60.0	25.5
D	79.3	24.4
E	67.2	30.3
F	46.5	32.9
G	39.7	41.8
H	31.3	37.4
I	27.6	34.0
J	25.5	36.3
K	28.6	33.7
Average	42.3	29.9
1 sigma	18.5	9.9

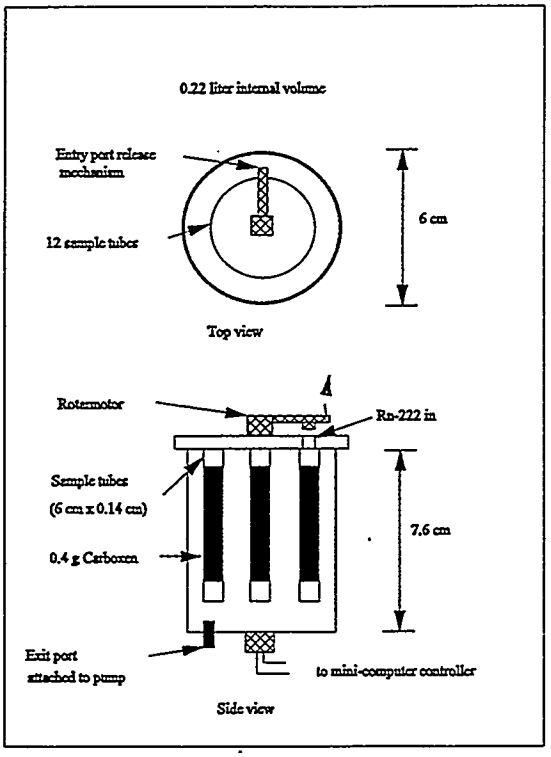

Figure 3.2 EML Polyport radon sampler.

References

Negro, V. C.
"Radometer- A Portable Field Instrument for the Rapid Measurement of Environmental Radon and Thoron"
IEEE Transactions on Nuclear Science, 37, 854-858(1990)

Polito, M. and B. Albert
"Low Power, Constant-Flow Air-Pump Systems"
USDOE Report EML-558, January (1994)

Aerosol Physics

3.11 EML/NYU EVALUATION OF A NEW PARTICLE SIZING TECHNIQUE

Keng-Wu Tu

A newly developed portable diffusion battery system was tested in the new EML radon chamber using Carnauba wax particles. The device was designed by Dr. Naomi Harley at New York University (NYU) and is being studied by her graduate student, Maire Heikkinen, as part of her Ph.D. thesis. This system was designed for long-term sampling at a flow rate of 2 L min^{-1} for measurement of indoor radon progeny particle size. The system utilizes conventional wire screens, as well as porous metal disks, to achieve separation of particles by size. Nuclear track detectors are used to record the activity penetrating each stage.

This system was tested against the EML radon progeny size measurement system, a combination of a graded screen array and a micro-orifice uniform deposition impactor (MOUDI), with Carnauba wax particles at sizes 70 and 100 nm (see EML Procedures Manual, in press). The radon concentration was maintained at 3400-4000 Bq m^{-3}.

The results are listed in Table 3.4. The agreement is reasonably good, but there is room for improvement.

TABLE 3.4

RESULTS OF EVALUATION

EML (^{218}Po)					NYU (^{218}Po + ^{214}Po)		
No. of samples	CN* (cm^{-3})	GAMD** (nm)	GSD+	%	Sampling time (h)	GAMD** (nm)	GSD+
3	11-17K	0.56 -0.72	1.2 -1.33	6-10	12	1-10	1.3-3
		94-98	1.3 -1.44	83-93		45-550	1.3-2.8
3	11-13k	0.64-0.82	1.31-1.44	19-45	6	1-2.6	1.3-1.8
		74-79	1.63-1.9	54-80		35-100	1.3-2.1

* Particle number concentration.
** Geometric activity mean diameter.
\+ Geometric standard deviation.

Reference

EML Procedure Manual
USDOE Report HASL-300, 28th Edition,
Section 2, in press

3.12 EVALUATION OF RADON PROGENY SAMPLER EFFICIENCY

Earl O. Knutson

To determine the overall efficiency of a radon progeny sampler, we must take into account the reduction of efficiency due to the deposition of particles in the sampler inlet, the collection efficiency of a particle filter, and the alpha counting efficiency of the detector. In this cooperative study with the Tae Soon Park[1], the efficiency of a certain radon progeny sampler (sampler A) was evaluated by using both theoretical and experimental methods. In the theoretical method, the inlet efficiency of the sampler was calculated using a standard aerosol-physics equation, yielding 0.2% for a particle size of 1 nm and 99.3% for a particle size of 100 nm. The collection efficiency of the filter used in sampler A was taken to be 100% based on information in the literature. The alpha counting efficiency for the geometry used in sampler A was calculated by using both numerical (Wilson and Short, 1988) and Monte Carlo methods, yielding 39.5% with both methods. Therefore, the theoretical overall efficiency for sampler A was found to be 0.08% at a particle size of 1 nm and 39.2% at a particle size of 100 nm.

In the experiment, sampler A was run inside the new EML radon chamber, with and without injection of aerosol particles, and reference samples were collected with an open-faced filter. The radon progeny deposited on an open-faced

[1] Visiting scientist from the Korea Research Institute of Standards and Science, Republic of Korea.

filter was measured by a ZnS scintillation alpha counter, and it was analyzed using the Thomas-Tsivoglou, Nazaroff, or Raabe-Wrenn method (Knutson, 1989). From the reference measurements, we calculated the number of atoms sampled by sampler A, and the consequent number of alpha particles. The overall efficiency for the sampler was then determined from the ratio of the counts measured by it to the number of alpha particles determined from the reference filter. The results were found to be 1.1 ± 0.2% at low particle concentrations with less than 10 particles cm^{-3} and 40.7 ± 2.0% at high particle concentrations with more than 10,000 particles cm^{-3}.

The agreement between theory and experiment was very good, especially at the higher particle concentration. The difference between theory and experiment at low concentrations can be explained as follows. In the theory, it was assumed that there are no particles larger than 1 nm in the chamber, but in fact there were a few. Thus, inlet losses were not as high as those predicted by theory.

From this experiment, we conclude that precautions must be taken when sampling radon progeny from air where the particle concentration is low.

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"Personal Computer Programs for the Use in Radon\Thoron Progeny Measurements"
USDOE Report EML-517 (1989)

Wilson, O. J. and S. A. Short.
"Geometric Efficiencies of Circular Detector and Surface Source Arrangements - Numerical Solution and Experimental Verification"
Nucl. Instr. and Meth., A271, 644-651 (1988)

3.13 DESIGN OF A NEW "MERCER CELL" SAMPLER FOR ULTRAFINE AEROSOLS

Alfred J. Cavallo

The dose rate to the human lung per unit of airborne radon progeny concentration is greatest for particle diameters between 0.5-10 nm, where it is quite difficult to measure particle size distributions with much precision. A better method to measure size distributions in this range would allow a significant improvement in the lung dose estimate. In addition, it would also advance the understanding of particle formation and nucleation.

The new instrument, a Mercer cell, designed at EML is based on measuring the diffusional deposition of particles from air flowing radially inward between two parallel surfaces (Mercer and Mercer, 1970). The deposition as a function of position along the surface can be calculated without the need for any experimentally determined correction factors. Since the deposition as a function of position along the surface can be measured, much more detailed information on the particle size distribution will become available. With current instruments based on using wire screens, only total deposition (or transmission) is measured. The deposition profile itself cannot be measured given the geometry of the screen.

Initially, measurements will be made using standard ZnS scintillator and photomultiplier tube techniques. Once the best geometry for the measurements is determined based on experimental and theoretical studies, a field-deployable version will be designed and built. In this instrument, the activity as a function of position can be measured in real time using a digital CCD (charged coupled device) camera. These devices are compact and rugged, and can easily be coupled to a computer for data acquisition and analysis.

Figure 3.3 shows the new Mercer cell. The air inlet is a very narrow slit (0.005 to 0.064 cm) in the circular knife-edge seen around the circumference. The Mercer cell was machined by the EML Instrumentation Division.

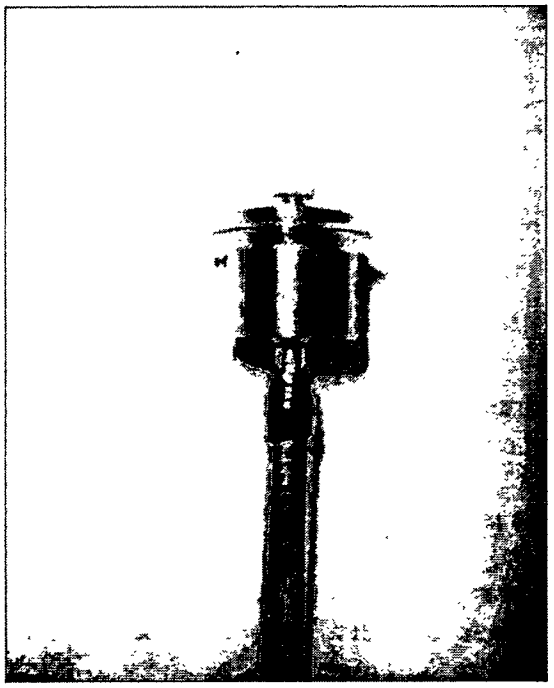

Figure 3.3 Mercer cell assembled.

Reference

Mercer, T. T. and Mercer, R. L.
"Diffusional Deposition from a Fluid Flowing Radically Between Concentric, Parallel Circular Plates"
Aerosol Sci., *1*, 279-285 (1970)

3.14 RECOIL LOSS OF ^{214}Pb FROM WIRE SCREEN SAMPLERS

Earl O. Knutson and Andreas C. George

An experiment was carried out this year to measure the loss of ^{214}Pb atoms from the surface of a wire screen due to recoil following the decay of atoms originally sampled as ^{218}Po. Recoil is a consequence of the large (6 MeV) energy release involved in the alpha-decay of captured ^{218}Po that is sufficient to drive the recoiling ^{214}Pb nucleus into the wire or into the air space between wires. In the latter case, any ^{214}Pb atoms swept away by air flowing through the screen are termed lost by recoil.

Samples were taken from filtered air containing about 4 kBq m^{-3} of radon gas, aged 2 to 3 min. Two sampling heads were used: an open-faced filter holder, and a holder containing a 39 mesh cm^{-1} wire screen with a backup filter. After sampling, a clean backup filter was installed and clean air was drawn through the screen for a set period of time. This backup filter was then alpha-counted. Simultaneously, the two original filters were alpha-counted and the amounts of ^{218}Po, ^{214}Pb and ^{214}Bi at the end of sampling were determined using our standard methods (George and Knutson, in press)

The results of four tests are shown in Table 3.5. These losses are quite small, and can be neglected in any of the work we do. This work was presented at the 1994 European Aerosol Conference (Knutson and George, 1994)

TABLE 3.5

RECOIL LOSS OF ^{214}Pb FROM A 39 MESH cm^{-1} WIRE SCREEN

Test No.	^{218}Po Activity on Screen (Bq)	^{214}Pb Recoil Loss*
1	31.8 ± 1.1	.043 ± .010
2	26.6 ± 1.3	.059 ± .013
3	28.5 ± 1.1	.059 ± .012
4	48.0 ± 1.5	.040 ± .007

* Maximum possible loss = 0.50

References:

George, A. C. and E. O. Knutson
"Raabe-Wrenn Least-Square Method"
EML Procedure Manual, HASL-300, 28th Edition, Vol. I, in press

Knutson, E. O. and A. C. George
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3.15 PLATEOUT DATA FROM MEASUREMENTS IN PENNSYLVANIA HOUSES

Earl O. Knutson, Gladys Klemic and Peter Shebell

The objective of this study was to review, analyze and tabulate data from the measurements of surface-deposited radon progeny activity made from August 1992 to February 1993 in four houses in Pennsylvania. Other data from this project, collected in conjunction with the Pennsylvania Department of Environmental Health and Porter Associates of Ardmore, PA, have been described in the two prior annual reports.

The sampling method used for the 1992-93 study was to mount 5 to 6 Whatman filter paper disks on representative surfaces in the houses. These swatches were left up for at least 3 h to allow the radon progeny radioactivity to reach equilibrium, then they were removed and quickly transferred to drawer-type scintillation alpha counters. Standard alpha counting was done to determine the activity on the swatches, and they were corrected to the time of removal.

Twenty nine of the data sets, with a total of 152 swatches, were of good quality. In 22 of these data sets, two of the swatches had been mounted side by side as a test of reproducibility. Using potential alpha energy as the measure of activity, the average relative difference between the paired swatches was 0.255. By comparison, the average relative standard deviation among the 5 or 6 swatches used in each test was 0.34, clearly larger than 0.255. Thus, from this data it is suggested that there are real differences among the different mounting locations.

The next step in this study will be to collect the above-described data with other data from the same houses into a consolidated database. This database will be used to check the validity of our computer model for indoor radon progeny aerosols, MIAQ_RNP (see Summary No. 3.16)

3.16 UPDATE ON THE MODEL FOR INDOOR AIR QUALITY, MIAQ_RNP

Earl O. Knutson

Work has continued on the computer code that predicts the amount and size distribution of radon progeny aerosol in a room of a house, given certain input data. As discussed in last year's annual report, this code was developed by Christian Lange from RISO in Denmark, while he was a visiting scientist at EML. The new code is a modification of the model MIAQ4 by W. W. Nazaroff, University of California at Berkeley.

After Christian Lange returned to Denmark in January 1994, he and Earl Knutson worked independently to test and make changes in the code. The two principals met in Denmark in June 1994 to address the differences that had developed in the code, and agreed on a list of 16 enhancements that they will continue to work on. Also, they settled on final name for the code MIAQ_RNP, which replaces the interim name M6RADON. The new name clearly shows the linage of the code, and shows that it includes a treatment of radon progeny.

Exposure Studies

3.17 PARTICLE SIZE MEASUREMENTS AT A FORMER URANIUM MINE (TWILIGHT MINE) UNDER SIMULATED MINING CONDITIONS

Earl O. Knutson and Keng Wu Tu

During May 17-23, 1994, scientists from EML, the USDOI Bureau of Mines, and Clarkson University conducted a unique air sampling project at Twilight Mine. The objective was to better document the characteristics of the air that was breathed by uranium miners in the 1950s and 1960s - information that is central to our present day knowledge of the linkage between radon and lung cancer. The strategy was to recreate some of the 1950s working conditions to sample the air using 1990s equipment.

Twilight, located about 160 km south of Grand Junction, CO, was an ore-producing mine from 1950-1971. In 1973, the Bureau of Mines leased the mine and equipped it to serve as a research and training facility. It continues to serve this function to the present.

Although no actual mining operations were conducted during May 17-23, the dust raised by the foot traffic of the 10 person sampling crew is believed to have simulated the dusty atmosphere to which the miners were exposed. Additionally, a small diesel ore-loading machine was operated in the mine. Bureau of Mines personnel indicated

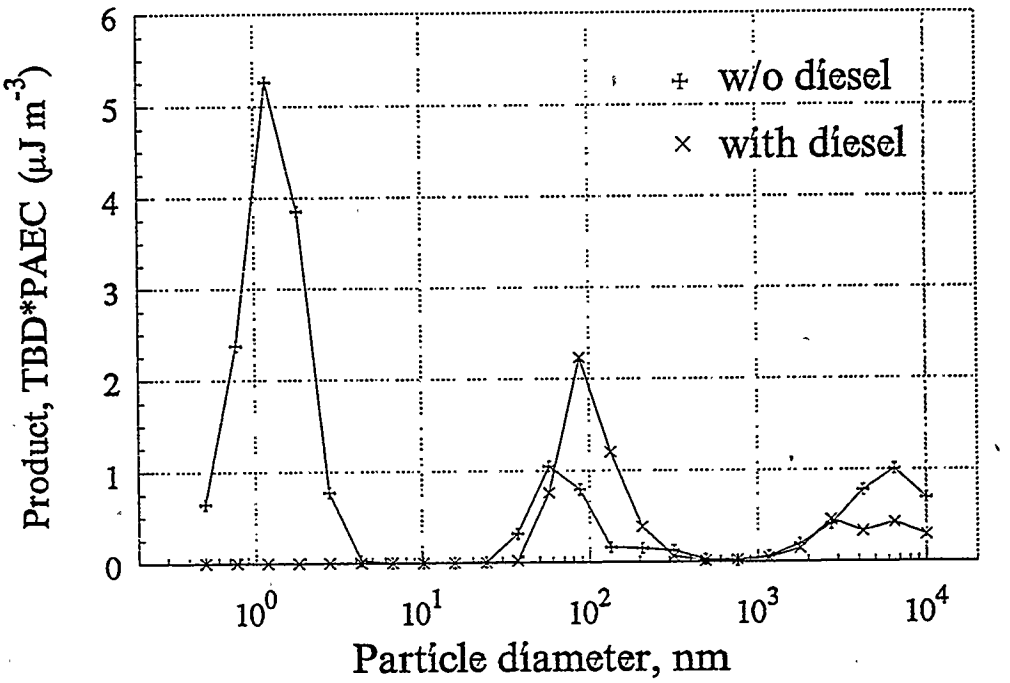

Figure 3.4 Contribution of various particle sizes to tracheobronchial deposition of radon progeny.

that such machines were commonly used in the mid-1900s.

An important finding from the new data is that particles 1 to 10 micrometers in diameter carry substantial amounts of radon progeny. Figure 3.4 shows the tracheobronchial deposition of potential alpha energy calculated from one test without diesel operation and one test with it. In both cases, a substantial contribution is seen from particles larger than 10^3 nm.

Current lung models show that these particles are very important in terms of radiation dose to the respiratory tract. It is expected that the data will be fully analyzed and reported by mid-1995.

3.18 FIRST VISIT TO A POTENTIAL SURROGATE URANIUM MINE - PEND OREILLE

Earl O. Knutson

The Pend Oreille Mine is located in the northeast corner of Washington State, about 5 km north of the village of Metaline Falls. The mine has produced zinc and lead since 1929, under several ownerships and managements. Under the current owner, Resource Finance, Inc., of Toronto, Ontario, Canada, exploratory mining is being done to determine the feasibility of resuming production mining.

Our interest in Pend Oreille is as a potential surrogate uranium mine. Radon levels in the mine are moderately high, and the type of mining activities are similar to those in the former uranium mines. If this analogy holds up under further study, a carefully designed experiment at Pend Oreille could illuminate on the exposure conditions found in the old uranium mines.

To begin exploring this possibility, Earl Knutson of EML joined a two-person team from the USDOI Bureau of Mines in a brief (October 25-27, 1994) radiological study of the mine. Exploratory mining was being conducted around the clock by a crew of 21 miners using the techniques of drilling, blasting, mucking and hauling. Large diesel machines were used for hauling. Transportation into and inside the mine is by means of diesel-powered pickup trucks. Figure 3.5 shows the sampling crew in one of the larger chambers of the mine, about 150 m below the surface.

In the survey, measurements were made of radon gas and progeny concentrations, gamma radiation, and the concentration and size of airborne particles. Most measurements were made at the D8 pump station, about 1 km from the area of active exploratory mining. Diesel ore-hauling machines were passing the station frequently.

The radon concentration at D8 was about 900 Bq m^{-3}. The particle concentration varied from 30,000 to 300,000 as the ore-hauling machines passed the station. Based on our present knowledge, we believe Pend Oreille is a good candidate for a surrogate uranium mine.

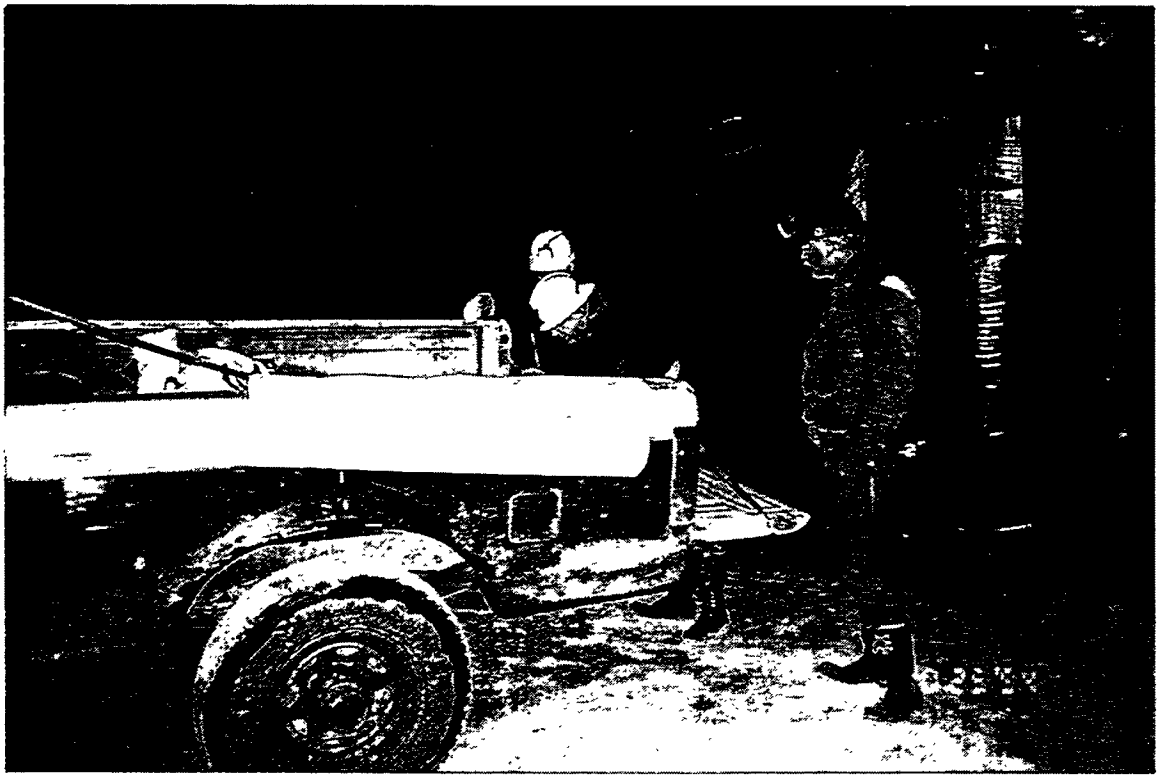

Figure 3.5 Sampling crew in a large chamber at Pend Oreille.

3.19 SOIL GAS ^{220}Rn AND ^{222}Rn

Adam R. Hutter

Soil gas ^{222}Rn is widely believed to be the major source of elevated indoor ^{222}Rn concentrations in U.S. homes and buildings. Much less is known concerning both soil gas and indoor ^{220}Rn, largely due to measurement difficulties arising from its short half-life of 55s. However, it is estimated that 10-20% of the total dose equivalent from all indoor radon is due to ^{220}Rn progeny. Ongoing studies at EML have been conducted in order to better understand the source variability of ^{222}Rn and ^{220}Rn, both temporally and spatially, especially geologic and geochemical controls. As part of these studies, soil gas ^{220}Rn and ^{222}Rn concentrations have been measured at two New Jersey sites at depths of 1.5, 1.0, 0.5, and 0.25 m on a bi-weekly basis for more than 1 y. Measurements at the 1 m depth have been conducted for more than 2 y.

One site at Chester, NJ, situated along the border of the Reading Prong and underlain by granitic gneiss, shows vastly different soil gas ^{220}Rn and ^{222}Rn characteristics than the other site located in Matawan, NJ, which is located within the coastal plain province and underlain by sands and gravel. The Matawan site ^{220}Rn and ^{222}Rn data show little temporal or horizontal spatial variability, but well-developed vertical diffusion profiles are observed in the ^{222}Rn data. At the Chester site, strong temporal and spatial variability, in both the vertical and horizontal directions, are present in the ^{222}Rn and ^{220}Rn data. Temporal changes in the ^{220}Rn concentrations behave according to diffusion theory as expected under both diffusion and advection conditions. However, the ^{222}Rn concentration variations at this site, up to 10X higher in the autumn than in late winter at all depths, are larger than can be accounted for by diffusion equations. The ^{220}Rn/^{222}Rn ratio data indicates that soil gas advection may be causing the highs in the ^{222}Rn concentration. Models compared with the profile ^{220}Rn/^{222}Rn ratio data show good correlation at the three shallower depths ($r \sim 0.8$), but not at the 1.5 m depth. Permeability measurements show very poor correlations with ^{220}Rn and ^{222}Rn concentration variations ($r < 0.2$).

These findings (Hutter, 1994) were presented at the 1994 Geological Society of America Annual Meeting. Additionally, a paper by Hutter (in press) describing the measurement method for soil gas ^{220}Rn, heretofore undocumented, was accepted for publication.

The direction of the soil gas ^{220}Rn and ^{222}Rn program in the future will be to focus on modeling the soil gas ^{220}Rn and ^{222}Rn data in an attempt to elucidate on the transport component of the observed seasonal variations.

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Health Physics, in press

3.20 INVESTIGATION OF POTENTIAL GAMMA DOSE FROM RADON PROGENY ATTACHED TO CLOTHING

Gladys Klemic and Earl O. Knutson

In response to a request from Georgetown University School of Medicine to assist in determining the potential exposure to gamma rays due to radon progeny attached to clothing, EML measured the activity deposited on various

surfaces under optimal plateout conditions to obtain an upper limit on the magnitude of the dose that could result. Concerns had been raised by a retired Georgetown breast cancer surgeon and a local Washington D.C. newspaper reported the hypothesis that enhanced progeny plateout on synthetic fabrics used in undergarments could be responsible for increased rates of breast cancer in women in the last 15 years.

In a previous study published by EML, it was found that radon progeny deposition was about the same for many materials (cotton, aluminum, paper, corduroy) but that polyethylene could collect two to four times more activity, perhaps due to electrostatic charge (Knutson et al., 1992). The same methods used in this previous plateout study were applied here with samples of various materials exposed under controlled conditions in the new EML radon chamber. Parameters were maintained for maximum plateout: high radon concentration (about 4900 Bq m^{-3} or 132 pCi L^{-1}), with low RH (17-21%), and very few aerosol particles (about 170 cm^{-3}). In one case, an attempt was made to impart an electrostatic charge to cloth samples (by rubbing them against another cloth and placing them on a CRT screen for a few minutes) before they were exposed in the radon chamber. Five samples were removed at a time to be counted simultaneously using windowless scintillators to detect alpha emissions in order to determine the surface activity of each progeny nuclide at the end of the exposure.

The results showed that while deposition could be higher by a factor of about 2 to 5 for materials deliberately charged, dose calculations by collaborators at Georgetown determined that the resulting annual dose to the breast is lower than that received in a single mammogram and is comparable to that due to natural background radiation. This study demonstrated that radon progeny deposition on clothing may result in measurable activity, but that the corresponding external radiation dose to the body would be insignificant, even for the relatively high radon concentrations used here which would be expected to result in a substantial internal dose. Therefore, it was concluded that radon progeny attached to clothing is not a factor in breast cancer.

Reference

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"Measurements of Radon Progeny Activity on Typical Indoor Surfaces"
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3.21 INDOOR RADON/THORON PROGENY MEASUREMENTS

Keng-Wu Tu

We made another seven indoor radon/thoron progeny measurement tests in 1994. This will conclude our indoor radon/thoron progeny study program for now. However, all the instruments related to this study will be maintained in good condition for any future needs. These seven houses are located in four different states, Colorado, Pennsylvania, New York and New Jersey. All of the houses had radon/thoron progeny PAECs < 50 nJ m^{-3} in the basements and the ground floors. The measurements, as in the past, were made with four Alpha Prism programmable radon/thoron progeny monitors manufactured by the Alpha Nuclear Corporation, Ontario, Canada.

The average measured basement and ground floor radon progeny PAECs were 41 and 33 nJ m^{-3}, respectively. The thoron progeny values were 14 and 22 nJ m^{-3}. As we found in our previous measurements, the ratios of the average thoron progeny PAECs to those of radon progeny in the low radon level homes were almost a factor of 2 higher in the ground floor than the basement. The results again indicate that building materials contribute significant thoron progeny activity indoors.

4

Atmospheric and Surface Pollutant Studies Related to Global Climate Change

4.1 OVERVIEW

Merrill Heit

Research performed in this program is diverse, encompassing several areas of atmospheric research. At present, the majority of the research is directed towards the role of atmospheric aerosols in processes related to global climate change or issues related to understanding the chemistry of the atmosphere. Studies involving the use of natural or anthropogenic tracers to investigate the transport mechanisms that distribute airborne pollutants within the atmosphere are also conducted.

During this past year, a significant portion of EML's climate research again focused on the development of an aerosol sampling system for the Atmospheric Radiation Measurements (ARM) Program's first Cloud and Radiation Testbed (CART) site. Both anthropogenic and natural atmospheric aerosol can have a profound effect on the earth's radiation budget. Aerosols can absorb or scatter radiation that can modify the earth's albedo and climate. Aerosols play a vital role in the formation of clouds by acting as cloud and ice forming nuclei. The size, morphology and chemical composition together with physical and dynamic processes determine whether the aerosol will be involved in cloud production or the earth's radiation budget. The source of these aerosols is from the earth's surface where they are transformed by chemical and physical processes into particles that may ultimately affect our climate.

Studies involving the characterization of aerosols in the boundary layer often require extensive aircraft support that is very costly and not always ideal for aerosol sampling. We have developed a prototype instrument package for installation on a light experimental aircraft operated by Oregon State University. This package has demonstrated the feasibility of using specially designed small aircraft for aerosol research.

EML is also currently using a global three-dimensional tracer transport model to examine the atmospheric transport of ^{210}Pb as an aerosol tracer produced by the radioactive decay of ^{222}Rn emitted from soils. The model results are compared with our global measurements of ^{210}Pb obtained from EML's Surface Air Sampling Program. The goal of this research is to improve

the simulations used in atmospheric modeling techniques such as convective and scavenging schemes, and turbulent boundary layer formulations used for tracer simulations. The improved capabilities will provide better simulations of the transport of aerosols and greenhouse gases related to global climate change.

Measurements of radon in the surface air at the National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostics Laboratory (NOAA/CMDL) site located at Mauna Loa, Hawaii, and at the Atmosphere Ocean Chemistry Experiments (AEROCE) site located at Tudor Hill, Bermuda, continue to provide information on the transport of continental aerosols to the oceans. The relationship between variations of radon and other atmospheric species such as ozone, ^{7}Be, ^{210}Pb, non-sea salt sulfate and nitrate is currently being studied in collaboration with the Department of Oceanography and Atmospheric Sciences, University of Miami (UOM), FL. The radon data will be useful for determining the origin of tropospheric ozone (stratospheric or urban). The meteorological processes and frequency of occurrence associated with the transport of continental air to these sites is also being investigated.

Prior to 1994, EML had also been active in the sampling and analysis of perfluorocarbon gases used as tracers in programs such as "Atmospheric Studies in Complex Terrain" (ASCOT) and the "Across North America Tracer Experiment (ANATEX). Some of these studies were still being completed in 1994. The purpose of these and related experiments is to validate models of atmospheric transport and dispersion of pollutants. EML's contributions in this area included improvement of the capillary-column chromatographic separation techniques, and the development of balloon-borne, light-weight, sampling systems for ascertaining the vertical distribution of the tracers within and above the atmospheric boundary layer.

In collaboration with the Air Force, a standby readiness capability is maintained by EML for balloon sampling in the atmosphere. The purpose of this readiness is primarily for documenting the injection of radioactive debris into portions of the atmosphere in the event of a satellite failure resulting in ablation of a nuclear reactor or radionuclide thermal generator (RTG) power source. On occasion, EML performs other experiments with the Air Force's balloon programs, such as measuring the vertical distribution of atmospheric aerosols from events like the eruption of a major volcano.

Collaborative efforts for the above projects are conducted with scientists located at the following institutions: Department of Forestry, Oregon State University, OR; Department of Oceanography and Atmospheric Sciences, University of Miami, FL; Crocker Nuclear Laboratory, University of California Davis, CA; the Department of Atmospheric Sciences, University of Hamburg, Germany; and NOAA's Atmospheric Research Laboratory, MD.

4.2 AEROSOL MEASUREMENTS AT THE ARM SOUTHERN GREAT PLAINS SITE: DESIGN AND SURFACE INSTALLATION

Robert Leifer, Ronald H. Knuth and S. Frederick Guggenheim

The Atmospheric Radiation Measurements (ARM) science team has decided that surface measurements of aerosols are needed at the Southern Great Plains site in Lamont, Oklahoma, because of the interaction of aerosols with long- and short-wave radiation (see Summary No. 4.3).

To improve the ARM Program radiation models, measurements of aerosol size distributions, condensation particle concentrations, aerosol scattering coefficients at a number of wavelengths, and the aerosol absorption coefficient are needed. In addition, continuous measurements of the ozone concentration are required. To meet the needs of ARM, EML has the responsibility to establish the surface aerosol measurements program at the Southern Great Plains site.

To provide representative aerosol measurements, EML designed a special sampling manifold described in last year's report. A stainless steel stack, having an inlet set at 10 m, provides ambient air to the aerosol sampling equipment located in a special trailer dedicated to aerosol measurements only. The air entering the aerosol instruments is dried by heating to a controlled temperature in order to reduce the relative humidity (RH) of the sample to below 40%. A separate sampling line made of Teflon, is co-located with the stainless steel aerosol sampling stack. This inlet is set at 10 m to provide ambient air to the ozone monitor. The stack and manifold should be ready for deployment and installation sometime in the fall of 1995.

ARM's operating protocol requires that all of the observational data be placed on-line, and in real time to be sent to the main computer facility. To accomplish this goal, Argonne National Laboratory is developing the data acquisition and control system. This includes both hardware and software interfacing. A field data ingestor (Sun Workstation) located in the aerosol trailer, will interrogate and control instruments via RS-232 serial interface ports. In addition, the field data ingestor will sample analog signals and perform control functions. Software programs will be written to assess the quality of the data before being sent to the ARM main computer facility for use by the ARM Science Team.

The final draft of drawings and statement of work (SOW) of the exterior support structure for the aerosol sampling stack was completed and submitted to the Site Program Manager. The sampling manifold was built and setup at EML for testing and interfacing to a Sun Workstation.

A new three wavelength nephelometer (λ = 450, 550, 700 nm) with backscatter capability (90° to 170°) has been purchased from TSI, Inc., and it was tested and incorporated into the existing aerosol manifold.

An adjustable relative humidity system is under development and together with the three wavelength nephelometer it will provide a measure of the scattering and backscattering coefficients as a function of RH and wavelength. A second single wavelength (λ = 550 nm) nephelometer (Radiance Research, Inc.) will be operated as a reference in one of the extra isokinetic sampling lines under dry conditions (RH < 40%). The results of last years research was reported in Leifer et al. (1994) and Lee and Leifer (1994).

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Proceedings of the third Atmospheric Radiation Measurement (ARM) Science Team Meeting, CONF-9303112, UC-402, pp. 349-351, March (1994)

4.3 AEROSOL MEASUREMENTS AT 60 METERS DURING THE APRIL 1994 REMOTE CLOUD STUDY/ INTENSIVE OPERATING PERIOD (RCS/IOP)

Robert Leifer, Brian Albert, Hsi-Na Lee,
Ronald H. Knuth and
Lambros Kromidas

In response to a request from Dr. Harvey Melfi of NASA for aerosol measurements during the April 1994 RCS/IOP, EML modified an existing EML aircraft aerosol sampler that had been previously tested (see Summary No. 4.5). The sampler was installed on the 60 m tower at the (ARM) Southern Great Plains site. A 2-hour battery operated in-strument package was modified into a 30 day automated package capable of sampling at 1 minute intervals. The package was returned to the ground daily for data downloading and calibration of an integrating nephelometer. Data was obtained from the integrating nephelometer ($\lambda = 530$ nm), a six channel optical particle counter, a condensation particle counter, pressure, temperature and relative humidity sensors, and mini-impactors. In addition, twice daily back trajectory analyses were calculated, which provided some insight into the origins of the air mass sampled.

In the middle of the experiment, two of the controlling computers were destroyed by lightning. The lightning caused loss of all but the integrating nephelometer data because it had its own internal computer set to record at a 15-minute average intervals.

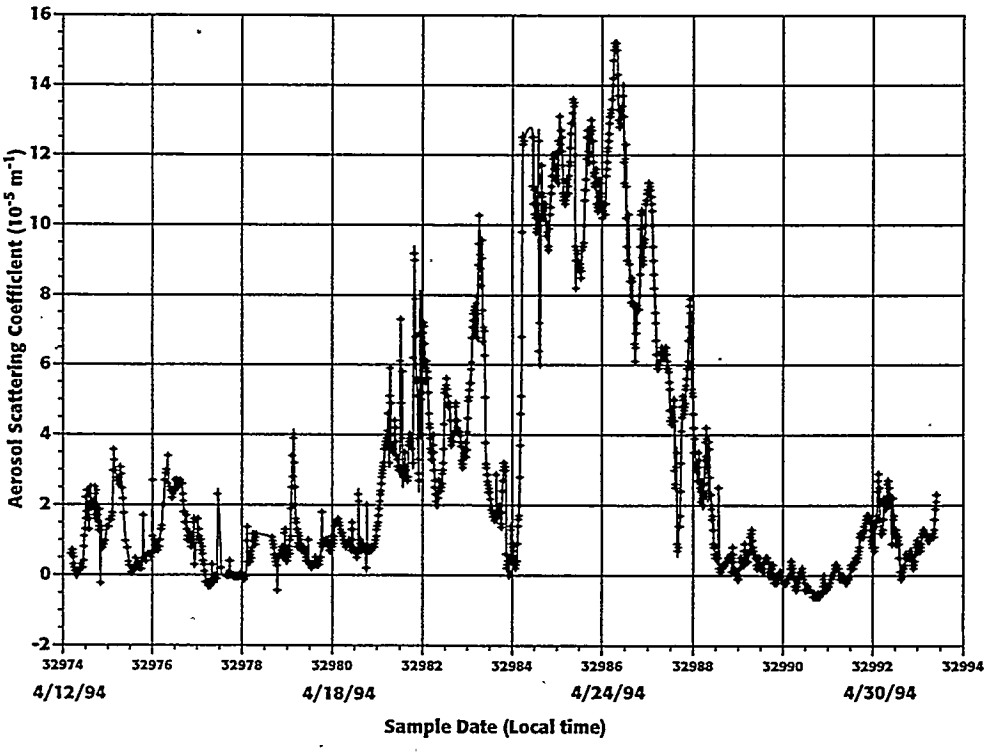

Figure 4.1 Aerosol scattering coefficient ($\lambda = 530$ nm) observations during the April 1994 RCS/IOP experiment at the site at Lamont, OK.

One of the reasons for the selection of the ARM Southern Great Plain site at Lamont, Oklahoma, is that many different types of air masses pass over the area. During the early period of the April RCS/IOP, extremely low aerosol scattering coefficients (Figure 4.1) were seen. This correlated very well with the back trajectory analysis which showed air coming from very clean sectors (continent and ocean). During the middle part of the experiment, extremely polluted air containing very high scattering coefficients impacted the site. The back trajectory analyses indicated the transport of air from the Illinois area during this period.

4.4 INNOVATIVE TECHNIQUES IN AEROSOL SAMPLING AND ANALYSIS

Robert Leifer, Lambros Kromidas, Ronald H. Knuth and Hsi-Na Lee

The vertical characterization of atmospheric aerosols was investigated using a high altitude balloon carrying a rotating drum impactor. The balloon was launched over Alamogordo, NM on February 12, 1992. At that time, we obtained a series of samples for scanning electron microscopy

TABLE 4.1

SUMMARY OF IMPACTOR SAMPLES

Sample ID	Sampling area characteristics	Comments
IS 1	• Sulfur found over entire impaction area • Large soil particles found in central impaction area • Sulfur, possibly as sulfuric acid, was the only element detected for aerosols below 0.2 μm • Zinc laden particles coincided with collected soil particles	• Low tropospheric • Impacted a plume with freshly produced aerosol • Smelter produced aerosols found in sample
IS 2	• Soil number concentration significantly reduced from IS1 • Zinc laden particles still seen in central core but at lower concentrations • Sulfur still associated with majority of particles • Lowest concentration of particles found in this sample	• Mostly upper tropospheric • Sulfur particles collected were bigger than IS1 and showed higher degree of volatilization
IS 3	• Dominated by sulfur and sulfur-silicon bearing particles • Sulfur particles primarily below 0.1 μm	• Lower stratospheric • Many of the sulfur-silicon particles had a central silicon core surrounded by sulfur
IS 4	• Dominated by sulfur and sulfur-silicon bearing particles • Size distribution of particles shifted to higher sizes, peaking at 0.3 μm	• Higher altitude (2 km) than IS3 • Sample reflects stratification of aerosol in stratosphere from Pinatubo
IS 5	• Sample collection of short duration but similar to IS3 and IS4	• Sample reflects both stratospheric and tropospheric collections

analysis. The aerosol samples were subsequently deposited on a substrate (carbon conductive adhesive; see Summary No. 4.6) with excellent collection efficiency. This substrate was found to be especially suitable for performing back-scatter microscopy and energy dispersive X-ray microanalysis. The following results were obtained for these samples:

---The entire vertical profile of atmospheric aerosols in our samples was dominated by sulfur.
---The low altitude samples provided information on the transport of zinc laden particles, possibly from smelters or mining in the Arizona, New Mexico or Mexican areas. Additional supporting evidence for the potential sources comes from back trajectory analyses, which showed trajectories of the air coming from Mexico and passing over Arizona before impacting on the Alamogordo area.
---The stratospheric samples contained large quantities of silicon laden particles indicative of the 1992 volcanic eruption of Mt. Pinatubo in the Philippines.

For this particular sampling day, the cleanest air (low number concentration) resided in the upper troposphere and not the stratosphere. This flight was terminated prematurely because of balloon failure and we were able to obtain only five samples. Table 4.1 provides a brief summary of the five samples. The results of this project will be submitted for publication in Atmospheric Environment.

4.5 FEASIBILITY STUDY: ENVIRONMENTAL SAMPLING ON A LIGHT EXPERIMENTAL AIRCRAFT (LEA)

Robert Leifer, Brian Albert and
S. Frederick Guggenheim

During the week of March 14, we tested an atmospheric environmental sampler on a light experimental aircraft (LEA; see last year's annual report). This was a feasibility study to deter-mine whether this small aircraft can be used as an inex-

Figure 4.2 Photograph of the LEA and instrument package used in the March 17, 1994 feasibility study at Corvalis, OR.

pensive and flexible platform for doing lower tropospheric atmospheric studies. A small sampler, weighing approximately 30 kg was mounted on the aircraft (see Figure 4.2). The sampler was automatically controlled by a computer, leaving the pilot to focus only on flying. Table 4.2 lists the major pieces of equipment included in the aircraft package.

Because of weather constraints, we were only able to fly two missions, one of 25 minutes duration and the second for 2 hours. Even though threatening weather conditions normally would have prevented most sampling flights from occurring during this week, the LEA allowed for two successful flights. Flights took off from a grass field close to the aircraft hanger. Typical air speeds for these flights ranged between 15 and 20 m s^{-1}, which is ideal for aerosol sampling. The site is located at an elevation of 313 m. In Figure 4.3 preliminary data are shown from an integrating nephelometer, condensation particle counter, ozone sonde, and pressure, temperature and RH sensors. A strong correlation exists between the condensation particle counter and the nephelometer, indicating that there was not significant submicron particles present below 0.1 μm. There are a few cases where the condensation particle count increased but the aerosol scattering coefficient did not, suggesting the possibility of fresh aerosol production.

Based on the preliminary analysis of the data, we found the LEA to be an extremely flexible aircraft, well-suited for boundary layer studies and extremely economic to fly. A journal article describing this flight will be available next year.

TABLE 4.2

INSTRUMENTATION USED ON THE LEA

Instruments	Manufacturer	Measurement
Intergrating Nephelometers	Radiance Research Model 901 Model 902	Scattering coefficients: at 470 nm at 530 nm
Condensation Particle Counter	TSI Inc.	Particle concentration greater than 0.01 µm
Optical Particle Counter	Met One, Inc.	Size distribution (0.3 to 5 µm)
Ozone Monitor	Astro Engineering, ECC Oxidant Meters, Model 002	Ozone concentration
Solar Radiometers	Licor LI-1990SZ	
Pressure, Temperature and RH Sensors		
Two Tattletale computers		

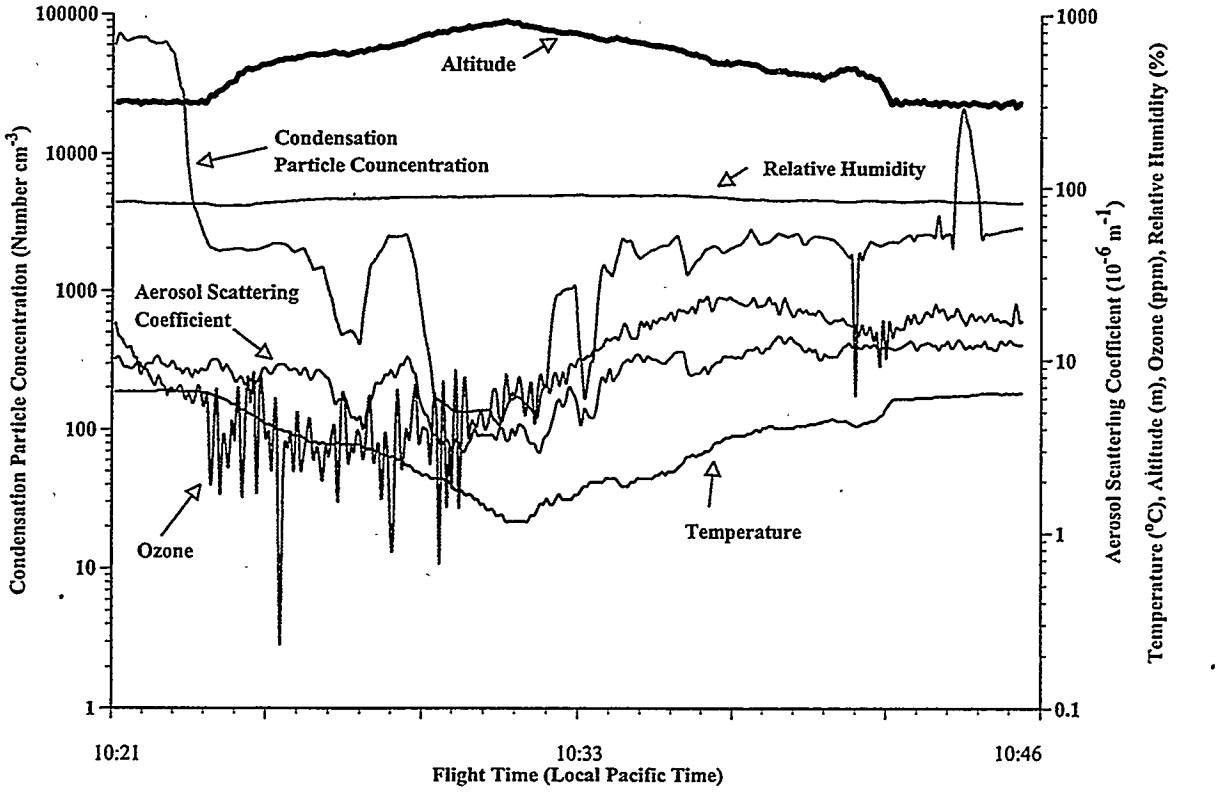

Figure 4.3 Preliminary results of the observations from the flight of a LEA on March 17, 1994 at Corvalis, OR.

4.6 USE OF A NEW TYPE OF IMPACTOR SUBSTRATE FOR COLLECTION OF AEROSOLS

Lambros Kromidas and Robert Leifer

We investigated a commercially available double-sided electrically conductive carbon adhesive for the collection of aerosols by impactors (Kromidas and Leifer, in press). This collection surface was found to be excellent for X-ray microanalysis since it does not contribute to the energy dispersive spectrum. X-ray microanalysis of this substrate showed it to contain only carbon and oxygen with carbon being the major component. This composition makes the carbon substrate ideal for backscatter electron microscopy because of maximal contrast differences between the substrate and collected non-carbonaceous particles. The use of this substrate enhances our capability to perform feature analysis and chemical characterization of aerosols by automated scanning electron microscopy.

Reference

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Atmospheric Environment, in press

4.7 ATMOSPHERIC MODELLING AND APPLICATIONS

Hsi-Na Lee

The following research illustrates EML's efforts to produce or modify atmospheric models which describe the mechanisms that distribute airborne pollutants within the atmosphere. Included are a three dimensional global tracer transport model (TM2) used for studying pollutant transport related to climate change and a semi-Lagrangian transport scheme with spectral interpolation for improving advective transport and diffusion of tracer.

Intercomparison of Wet Precipitation Scavenging Schemes and the Emission Rates Of ^{222}Rn for Simulation of Global Transport and Deposition. We simulated the transport of ^{222}Rn and its progeny ^{210}Pb using a global three-dimensional atmospheric tracer model. The purpose of this study was to understand the effects of wet deposition scavenging processes and the emission rate of ^{222}Rn on the global transport and deposition of ^{210}Pb. We examined the deposition of ^{210}Pb aerosols due to wet scavenging in stratiform and convective precipitation. Four schemes for wet precipitation scavenging removal, and two assumed distributions of the emission rate of ^{222}Rn were studied and tested. One distribution was assumed to have a global constant rate and in the other the constant rate was assumed to have changed with latitude. The results of the model simulations were compared with archived radionuclide data measurements from EML's global network of sampling stations. The model-calculated global-distributions of the yearly mean of the surface air concentrations of ^{222}Rn and ^{210}Pb, and the yearly mean total deposition of ^{210}Pb for 1986 data for each wet precipitation scavenging scheme studied are presented. For each scheme, we also compared yearly mean total deposition and monthly averages of the surface air concentrations and the vertical distributions of ^{210}Pb with the measurements. Figure 4.4 shows each scheme calculated for the global average of monthly bias and the RMSE of ^{210}Pb surface air concentrations that deviated from measurements obtained from EML (Lee and Feichter, in press). The model comparison with the measurements helped us to validate and improve the model. This atmospheric tracer transport model can be used for the simulation of other radionuclide and nonradionuclide tracers for future studies related to global climate change.

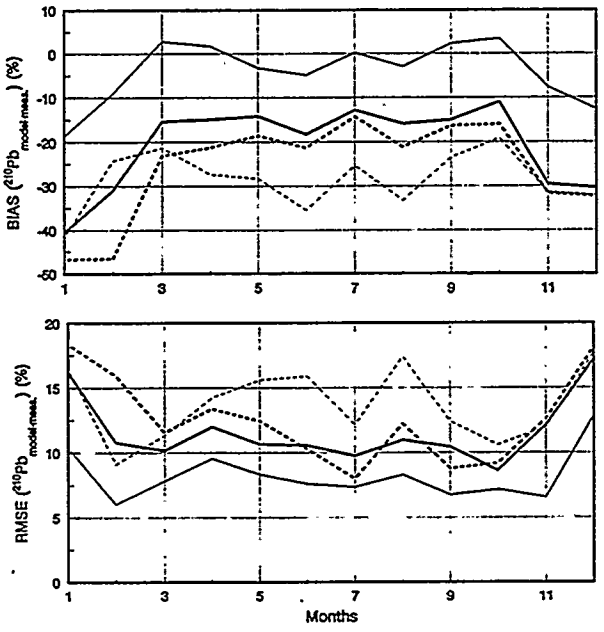

Figure 4.4 Model-calculated global average of monthly bias and root-mean-error of ^{210}Pb surface air concentrations for each scheme.

A Transport and Diffusion Model Using the Semi-Lagrangian Technique with the Spectral Method. Many researchers have devoted considerable effort to understand the transport and diffusion mechanisms of atmospheric pollutants. An atmospheric transport-diffusion equation has been widely used by scientists for modeling the behavior of pollutants in the atmosphere. To calculate the pollutant transport and

diffusion, the equation is solved by a numerical method. The most popular method is to discretize the equation based on a finite-differencing technique. A large number of finite-differencing techniques have been developed to reduce the numerical errors by solving the atmospheric transport-diffusion equation. Other numerical techniques that offer an alternative approach, such as spectral and pseudospectral techniques, can minimize diffusion errors. The semi-Lagrangian technique is also used successfully by researchers for studying advective transport. A technique was developed which coupled the semi-Lagrangian technique with the spectral method for solving the advective equation (Lee, 1993). This technique has been extended in this present work to solve a transport-diffusion equation (Figure 4.5). By utilizing this technique for modeling pollutant transport and diffusion, the distributions of pollutants under various atmospheric stabilities have been calculated.

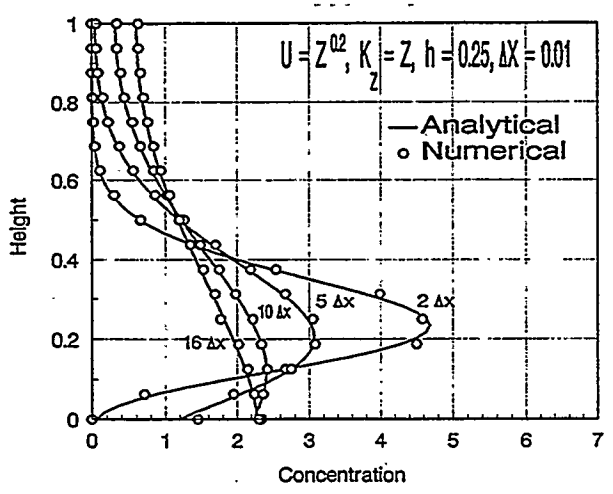

Figure 4.5 Vertical profiles of the steady-state solutions of a transport-diffusion equation in a non-dimensional form for a point source at height, h = 0.25. Grid points 17 x 17 with a grid spacing of $\Delta x = 0.01$ were used.

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4.8 ATMOSPHERIC RADON MEASUREMENTS AT BERMUDA AND MAUNA LOA, HAWAII

Adam R. Hutter and Richard J. Larsen

EML's ^{222}Rn analyzers, deployed at the Mauna Loa Observatory, operated under the auspices of the National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostics Laboratory and at the Atmospheric/Ocean Chemistry Experiment Site, at Tudor Hill, Bermuda, continued in 1994 to provide reliable ^{222}Rn data. These data are used to determine air mass provenance, specifically, the timing of continentally-derived air transported to the mid-Atlantic and mid-Pacific. Additionally, the ^{222}Rn data are used to better understand local meteorological effects, as well as to provide the scientific community with a rare database that can be used to validate global transport models.

In 1994, Adam Hutter highlighted some uses of atmospheric ^{222}Rn data at the American Nuclear Society's Third International Conference of Methods and Applications of Radioanalytical Chemistry in Kona, HI. Additionally, a paper by Hutter et al. (in press) identified some local and distant sources and the transport of ^{222}Rn to

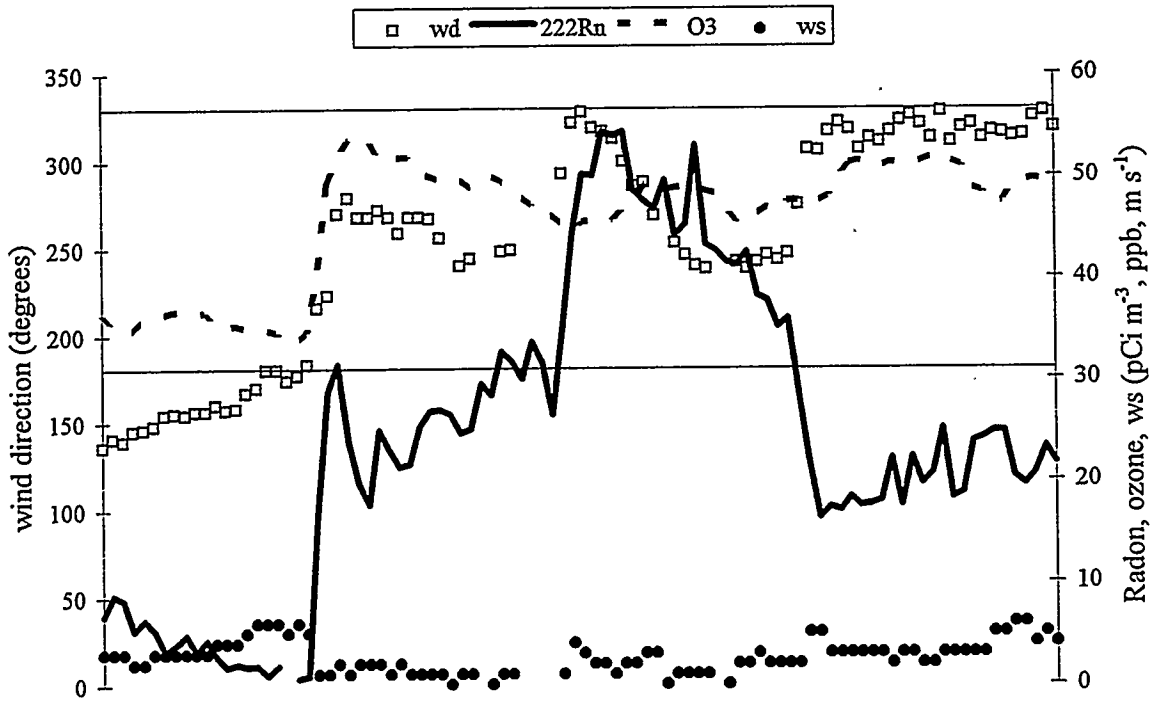

Figure 4.6 Data indicating arrival of continental air at Bermuda, noted by increased 0_3 and ^{222}Rn (wd = wind direction, ws = wind speed).

EML's Bermuda and Hawaiian sites. Figure 4.6 shows the usefulness of ^{222}Rn data in documenting the arrival of continental air, in this instance to Bermuda. A report detailing the results of the First International Intercomparison of Atmospheric ^{222}Rn Analyzers was provisionally accepted for publication in the Journal of Geophysical Research (Colle et al., in press).

The reliability of EML's ^{222}Rn analyzer and the utility of the resulting data to the scientific community has spurred requests for additional ^{222}Rn analyzers to be constructed. In the future we plan to expand the surface measurements of atmospheric ^{222}Rn to meet the growing needs of the scientific community. Towards this end, a new generation of EML's ^{222}Rn analyzers, based on electrostatic collection of ^{218}Po, is being developed (see Summary No. 5.5).

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J. Radioanal. Nuclear Chem., in press

4.9 ATMOSPHERIC TRACER STUDIES

Raymond J. Lagomarsino

The analyses of vertical and surface samples collected in Dolan Springs, Arizona during MOHAVE (Measurement of Haze and Visual Effects) has been completed. MOHAVE was a tracer experiment designed to determine if pollutants originating from El Centro, California, the Los Angeles basin and a coal-fired power plant located in Laughlin, Nevada are transported to Grand Canyon National Park (GCNP). These pollutants are thought to contribute to the park haze problem. A different perfluorocarbon tracer was released from each location.

Vertical samples were collected with a 7.25 m^3 balloon and a multiple-port sampler (Polyport Polito and Albert, 1994) attached to the balloon tetherline to determine the tracer concentrations at different altitudes up to 500 meters above the surface. A Polyport, weighing approximately 850 grams, contains a sampling pump, batteries, 12 adsorbent tubes and a microcomputer with associated electronics. The on-board computer provides: on/off pump operation, indexing of the Polyport sampler, measuring and storing sensor readings (temperature, pressure and RH), and control of the timing of all operations.

Vertical samples were analyzed by a newly developed gas chromatograph capillary column system utilized for the surface samples. This system not only improves the separation of the tracers of interest but also enhances the limits of detection by factors of two to five. The results of all of the analyses are presently being compiled and evaluated for inclusion into a final MOHAVE report.

Reference

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"A Low Power Constant Air Flow System"
USDOE Report EML-558, January (1994)

5 The Atmospheric Chemistry Program

5.1 OVERVIEW

Merrill Heit

Historically, this program has had two major components: "Pollutants in Deposition and the Biosphere"[1] and "Environmental Isotope Geochemistry." Due to changing priorities, this program was not funded by the Department of Energy's Office of Health and Environmental Research (OHER) in 1994 and was replaced by research in the area of atmospheric chemistry. Some of the research described as "Environmental Isotope Geochemistry" has continued and is described in the Environmental Radiation and Radioactivity program area.

EML now performs research for the Atmospheric Chemistry Program (ACP) in several areas that are necessary for long-range planning in fulfillment of the National Energy Policy Act. This research includes five components described below:

1. "Development of a Computer Database from EML's Sampling Programs and Project Stardust": The objective of this work is to provide a comprehensive computer generated database to the scientific community that will incorporate all published data from relevant EML programs and a complete characterization of EML's archived samples.

2. "Trans-Pacific Transport of Combustion Products from Asia": The objective of this component of the program is to evaluate, using modeling and measurements, the magnitude of the trans-Pacific transport of Asian combustion products to the U.S.

3. "Aerosol Characterization on Gulfstream Aircraft Samples": The purpose of this project is to provide and operate an aerosol sampler which will produce independent evidence of whether air parcels sampled in NPACS Grumman Gulfstream-1 (G-1) flights have a marine or continental origin. This project will also provide an independent verification of whether a fraction of the air intercepted during G-1 flights has a recent stratospheric origin. Additional measurements

[1]Formerly the Ecological and Subsurface Pollutant Research Program

will be made in support of a 3-year program over the Western Atlantic Ocean to understand halogen chemistry on ozone in the marine boundary.

A related component of this program is to measure radon and/or radon progeny during ACP G-1 campaigns. These data will be used to indicate or verify the presence of continental air on a real-time basis during flight.

4. "Radon Source Term to the Atmosphere": The purpose of this project is to discern and quantify the parameters controlling the regional variations of radon fluxes. These data will be incorporated into the generation of a global map of the radon source term to the atmosphere values for validation studies of global climate models.

EML is also actively engaged in collaborative efforts with members of the ACP scientific community, such as Brookhaven National Laboratory, Pacific Northwest Laboratory, ENVAIR (Kenniwick, WA), as well as with scientists located at the University of Miami, New Mexico Institute of Mining and Technology, Yale University, University of Alaska in Fairbanks, the Max Plank Institute for Meteorology, Hamburg, Germany, the U.S. Geological Survey, National Oceanic Atmospheric Administration's (NOAA), National Geophysical Data Center, Climate and Meteorology Diagnostics Laboratory (CMDL), the National Center for Atmospheric Research, the NASA/Goddard Institute for Space Studies, the University of Washington, the U.S. Army (White Sands, NM), and the U.S. Navy's Oceanographic Data Center.

5.2 EML SAMPLE ARCHIVES

Robert Leifer, Camille Marinetti and Nita Chan

In recent years, there has been a need in the atmospheric modeling community for a complete computer database of all the stratospheric and upper tropospheric radioactivity measurements. At the present time there does not exist a usable database encompassing the vast number of measurements of radionuclides made between the years 1957-1993 in the troposphere and lower stratosphere. Almost 50% of the data is published in reports that are not readily available to the scientific community, nor is it in a form that can easily be used. Only EML, because of our historical involvement and first-hand knowledge of the filter samples, can develop the complete database in a format that can be used by present day atmospheric modelers. These data are useful for development and verification of large-scale transport and climate models. These data can also be used for understanding tropospheric and stratospheric transport processes, and for helping model the future atmospheric impact of a projected new fleet of stratospheric flying aircraft.

To this end, EML has completed phase 1 of the stratospheric radionuclide database (RANDAB) program. This computer program represents the world's largest collection of stratospheric and upper tropospheric radionuclide data ever compiled for computer analysis. These data represent measurements obtained from Projects ASHCAN, STARDUST, AIRSTREAM and the High Altitude Sampling Program (HASP) for the years 1957 through 1983. More than 50,000 filters were collected during this period and

analyzed for up to 20 different radionuclides. All the available data associated with each filter is included in the database. We expect to have RANDAB on-line late next year.

In addition to the radionuclide database, EML's stratospheric database of trace gases, collected during Project AIRSTREAM has been completed and is presently available through EML. This database contains information on more than 1000 samples. Each sample was analyzed for one or more of the following gases: CCl_3F, CCl_2F_2, CCl_4, N_2O, F_6, CO_2, CH_4, CH_3CCl_3, and COS. The trace gas database has been transferred to the Carbon Dioxide Information Analysis Center (CDIAC) for national distribution.

5.3 TRANS-PACIFIC TRANSPORT OF COMBUSTION PRODUCTS FROM ASIA IN THE SURFACE AIR

Richard Larsen, Hsi-Na Lee, Robert Leifer and Matthew Monetti

A study was conducted to determine the feasibility of establishing sampling sites on islands in the North Pacific Ocean or along the Northwestern Coast of North America that would be suitable for measuring and documenting Asiatic pollutants traversing the North Pacific Ocean. Trends in the emissions of SO_x indicate that Asian emissions have nearly doubled between 1970 and 1986. The impact of increased Asian emissions not only can affect the U.S. directly, the emissions can also affect the oxidative and radiative properties of the atmosphere. Nine existing EML surface sites (Cold Bay, Alaska; Shemya Island, Alaska; Estevan Point, British Columbia; Cheeka Peak Observatory, Washington; Cape Meares, Oregon; Midway Island; Okinawa Island; Minamitorishima Island; and Mauna Loa Observatory, Hawaii) were identified that are potentially suitable for this study. The sites located at Okinawa, Midway and Shemya seemed to be the best available pelagic sites to intercept Asian pollutants in transit. However, it has been demonstrated that the bulk of the transport occurs above the marine boundary layer, and that surface measurements may not be beneficial in defining this transport. Therefore, EML's current role in this project will be to assist the ACP by evaluating, using modelling and available measurements, the magnitude of the trans-Pacific transport of Asian combustion products (see Summary No. 5.4). It is possible that surface measurements of radon at Shemya Island may be initiated, depending on the modelling results and the results from the measurements of ozone and carbon monoxide at Shemya that are currently being made by the University of Alaska in Fairbanks.

5.4 MODELING TRANS-PACIFIC TRANSPORT OF COMBUSTION PRODUCTS FROM ASIA BY USING THE GchM MODEL

Hsi-Na Lee

As stated in Summary No. 5.3, the transport of Asian air masses across the Pacific Ocean has recently attracted the attention of the atmospheric sciences community. In addition, Asian fossil-fuel consumption and emissions are expected to continue to rapidly grow. The objective of this research is to study and document the current and future trends of trans-Pacific transport of combustion products from Asia, and to understand the potential impact of Asian pollutants to the U.S. continent. A three-dimensional global chemistry model (GchM), which was derived from the original code (PLUVIUS) developed by Dr. J. Hales, ACP scientific team leader and advisor, was selected by EML to quantify and understand this potential impact. The model applies a Eulerian frame and is capable of simulating

complex chemistries and other source-receptor behavior of any specified number of pollutants.

In 1994, joint modeling efforts between EML and Dr. J. Hales were initiated to examine the performance of GchM for ^{222}Rn transport across the Pacific Ocean. These efforts are continuing and plans for additional extensive modelling simulations are being prepared.

5.5 AEROSOL CHARACTERIZATION ON DOE'S GULFSTREAM AIRCRAFT

Robert Leifer, Lambros Kromidas and Brian Albert

Sea salt particles are continually injected into the marine boundary layer. Most of the chlorine from these aerosols are redeposited back to the Ocean. A fraction of the chlorine from the salt particles is liberated as gaseous inorganic chlorine (up to 20%). The mechanisms by which the chlorine is introduced into the marine boundary layer, i.e., particle to gas conversion is not fully understood. Few published research studies have provided information on the principle reactants and the product species. The DOE Atmospheric Chemistry Program has developed a 3-year program focusing on halogen and ozone chemistry in the Marine boundary layer of the western Atlantic Ocean. One aspect of this program is EML's responsibility to develop the instrumentation necessary to evaluate the chemical speciation of aerosols found in conjunction with measurements of halogenated species on the G-1 aircraft.

EML will be using a recently developed rotating drum impactor designed specifically for use with a scanning electron microscope (SEM) to evaluate the aerosol chlorine deficits as a function of particle size in the marine boundary layer. This impactor, known as 4-PI or parallel impactor, has the capability of collecting up to four simultaneous aerosol samples. Controlled by a small computer, the impactor can sequentially collect more than 400 samples on a single drum for SEM analysis. The samples can be preset for collections at specific times or driven by any independent instrument, having an analog output, such as a pressure transducer for measurements in and out of the marine boundary layer or radon monitor for looking at continental air. Samples collected by the 4-PI will be analyzed using EML's SEM and X-ray analysis capability.

During this year major strides were made in developing the stage automation package for the SEM. Working with PGT, the manufacturer of the X-ray analysis system, we can now automatically analyze a sample and obtain both size distribution and elemental information. The system randomly selects a series of areas to analyze within the impaction area. It continues to collect data until it reaches a preset number of particles at which time it generates a series of reports on the sample. These are automatically saved in the computer for further analysis. Since the location of each particle is recorded, we can go back to the same particles for more analyses. The intent of the stage automation package is to develop an integrated system for rapid throughput of samples (see Summary No. 5.6).

5.6 MEASUREMENTS OF RADON AND RADON PROGENY USING THE ACP RESEARCH AIRCRAFT

Vincent C. Negro, Richard Larsen and Harold L. Beck

Systems to measure both charged and total ^{218}Po, and to collect ^{222}Rn, have been designed and developed at EML for operation on the Grumman Gulfstream 1 (G-1) research aircraft. These systems described below will be in operation during the DOE ACP Ozone Production Study

planned for 1995-1997. The measurements will be used to indicate or verify the presence of continental air on a real-time basis during the flights for decision making purposes. Estimates of the relative age of air masses may also be inferred from the radon data. Radon is a very sensitive indicator of continental air masses since the continents are strong sources of radon while essentially zero radon is released from the oceans.

Radgrabber. A new EML instrument, called the "Radgrabber", is capable of making measurements of charged ^{218}Po. Because of the short half-life of ^{218}Po (3.05 min), its concentration should be in close equilibrium with that of its parent ^{222}Rn, especially in the free atmosphere. Although not all of the ^{218}Po is charged, the charged fraction in the free troposphere should be relatively high except under conditions of high absolute humidity. The principle of the Radgrabber is based on the use of a strong electrostatic field to collect positively charged ^{218}Po atoms directly on a solid-state alpha detector. As the ^{218}Po decays, the detector and the associated electronics spectroscopically measure and register this information. The electrostatic field is established by maintaining the instrument's alpha detector, preamplifier, pulse height analyzer and Tattletale computer at high voltage (15-30 kV) with respect to the grounded outer case. Communication with the analyzer while the Radgrabber is in operation is achieved by using optical isolation. The sensitivity of the instrument is dependent on the air flow which can be quite high since there are no filters or restrictions to flow. However, as the velocity of the air increases a limit is reached beyond which the electric field can no longer attract the charged ^{218}Po to the detector. In the present instrument with a high voltage of 25 kV, the velocity limit corresponds to a flow of 40-50 scfm. At altitude, because of the rarefied air, this will reduce to 20-25 scfm. Under this condition, a concentration of 10 pCi/m^3 (.37 Bq m^{-3}) theoretically yields 50-75 counts in a 10- min interval. The Radgrabber will make precise measurements of charged ^{218}Po with a time resolution of 10 minutes and a lower limit of detection of less than 10 pCi/m^3 (.37 Bq m^{-3}).

Radon Gas Sampler. For measurements of ^{222}Rn, grab samples will be obtained using a radon gas sampler instrument. The instrument consists of two sections that are joined together for the flight. In one section are eight charcoal traps in a dry ice container, while the other section contains rotary and solenoid valves, a mass flow controller, a vacuum pump, and a data acquisition computer. During the flight, the operator initiates a sample cycle by selecting the next available trap with a manual lever and then pressing a start button. From this point on, the operation is automatic: the computer operates solenoid valves, the vacuum pump and the mass flow controller in the appropriate sequence and then measures the actual mass flow for the 10 minute sample time. The computer then performs a shutdown sequence and waits for the next sample initiation. After the flight, the manual valves on each trap are closed and the two sections of the instrument are disconnected, this allows removal of the individual traps. The traps will be immediately air-freighted back to EML where the ^{222}Rn will be de-emanated into and counted in the EML radon pulse ionization chambers (PICs). The traps will then be regenerated and air-freighted back for use on subsequent flights. The EML PICs are among the most sensitive low-level radon measurement systems in the world and their use in conjunction with the cooled charcoal traps will allow a minimum detection limit of about 0.2 pCi (.0074 Bq). This combination should allow concentrations of less than 5 pCi/m^3 (.18 Bg m^{-3}) to be measured with a 10 minute sampling time. The exact detection limit will depend on the time required to return the traps to EML and the exact mass flow rate at a given altitude. Since only a limited number of charcoal canisters are available, only about 24 samples per week can be processed.

However, these should be sufficient to validate the Radgrabber data over a wide range of experimental conditions.

5.7 RADON SOURCE TERMS TO THE ATMOSPHERE

Adam R. Hutter and Carl V. Gogolak

Currently, a single-value global-average of 1 radon atom cm^{-2} sec^{-1} emission rate as the source term to the atmosphere is used to test and validate the transport component of global climate models. A more realistic source term is needed in order to more effectively utilize and apply these models. The goal of this project is to produce global sets of radon source terms to the atmosphere that characterize regional and temporal variations. Initially, the project plan was to simply compile the available ^{222}Rn flux data (or similar data, such as aerial gamma surveys) and piece together a world map of radon source terms to the atmosphere. Towards this end, a comprehensive literature search for reports of radon flux data and relevant studies (e.g., surface radium data, aerial gamma survey data) has been conducted and a database compiled. However, these data are insufficient to directly produce global maps of the ^{222}Rn source term to the atmosphere due to limited geographic coverage, as well as the problem of bringing into register the data obtained using different techniques measured under a myriad of environmental conditions performed by a plethora of researchers. Therefore, a new approach to produce a global map has been implemented, in conjunction with researchers at the New Mexico Institute of Mining and Technology that involves the development of a model for ^{222}Rn flux that incorporates soil moisture, soil ^{226}Ra, and soil texture components. The data, which will be put into the model to produce 1° x 1° maps of ^{222}Rn flux to the atmosphere, are obtained either directly or via surrogates from existing global databases available from the U.S. Geological Survey, the NOAA/National Geophysical Data Center, the National Center for Atmospheric Research, the NASA/Goddard Institute for Space Studies, and other institutes. The existing data sets on ^{222}Rn flux, mainly from the U.S. and Australia, will be used to "calibrate" the ^{222}Rn flux maps produced from our model using the global databases of soil properties.

One of the many problems encountered in producing the ^{222}Rn flux maps was in the dearth of knowledge concerning seasonal variations of the ^{222}Rn flux, specifically the effect of surficial freezing and snow cover on the ^{222}Rn flux. Towards this end, a sampling project in a homogeneous setting (coastal plain province sands and gravels) was initiated to study this effect, as well as the overall reproducibility and quality assurance of ^{222}Rn flux measurements using the accumulation technique, the most commonly used method. The measurement protocol in this project uses four accumulation cans (A ~ 640 cm^2; V ~ 9300 cm^3) placed as near to one another as possible. First, the background ^{222}Rn concentration is determined, then the concentration that has subsequently built up after typical collection times of 30 min. The flux is determined from this ^{222}Rn concentration difference, the can dimensions, and the time. Four sets of these measurements are obtained, giving 16 samples in a 2 hr period. Due to this small time difference, environmental conditions are assumed to be constant for a set of 16 samples. A second set of 16 measurements is often obtained on the same day four or five hours after the end of the first set, and these are evaluated separately, since conditions may have significantly changed to affect the ^{222}Rn flux, e.g., temperature and barometric pressure. In addition, soil moisture, soil temperature and outdoor temperature are concurrently measured.

In summary, the percentage error in the ^{222}Rn flux measurements, determined from the ratio of

the arithmetic mean of the standard deviation to the arithmetic mean of the ^{222}Rn flux for each set of measurements was determined to be ~25%. Approximately half of this value is attributed to the error in the ^{222}Rn measurement, estimated to be 10-12% at the low concentrations measured. These findings, and their effect on the usefulness of the available data in the literature collected using similar methods that may be used to "calibrate" the global source term data sets, were presented at the 1994 Fall Meeting of the American Geophysical Union (Hutter and Gogolak, 1994).

The data obtained from the ^{222}Rn flux measurements will be used to determine the soil moisture and temperature dependence in the model calculations of ^{222}Rn flux from existing global databases (e.g., soil texture, soil depth, temperature, soil water potential, etc.). Once these correlations are determined, sets of global ^{222}Rn source terms to the atmosphere will be produced for each season and made available to the scientific community through journal publication(s). Additionally, intercomparisons and intercalibrations of experimental methods with other researchers, including those at New Mexico Institute of Mining and Technology and Yale University, are scheduled to take place in 1995 immediately following the NRE-VI symposium in order to improve QA in this area.

Reference

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EOS, Vol. 75, No. 4 (1994)

6

Metrology, Consultation, and Emergency Response

6.1 OVERVIEW

The EML Metrology program consists of developing the generic instrumentation and analytical methods required to improve our overall measurement and analysis capability. It also includes a variety of quality assurance efforts designed to further the state of the art of both EML's research, as well as environmental research carried out at other institutions. Also included in this section are projects associated with maintaining and improving the Laboratory's special facilities, such as the VAX Computer Center, the High Altitude Test Chamber (HATCH), the Instrumentation Division Machine Shop, and the Chester, NJ, field station. These facilities support many different EML research programs.

Because of the accumulated expertise and high reputation of the EML staff, we are frequently called upon by other agencies and institutions to provide advice and/or short term assistance on potential or actual environmental contamination problems. We also are occasionally called upon to provide assistance in matters involving emergency response to environmental contamination. The major consultation activities which we participated in during 1994, which include important interactions with the Nuclear Regulatory Commission (NRC) and the International Atomic Energy Agency (IAEA) are also discussed in this section. There were no significant emergency response activities during 1994.

6.2 INSTRUMENTATION DESIGN, DEVELOPMENT AND SUPPORT

Instrumentation Division Staff

The Instrumentation Division continued its role of providing design and support services for EML field and laboratory projects during 1994. This support included technical and design advice from the engineering staff, procurement of special equipment and supplies, aid in maintenance of facilities and computer links, as well as repair and maintenance, instrument modifications, and equipment assembly by the machine shop and technical services staff. Since the aforementioned support spanned most of the Laboratory's programs and are thus discussed in other sections of this report, only a few of the more significant efforts are listed below.

- Alternative Radon Detection Techniques: An investigation was begun into a new flow-through spectroscopy instrument where sample air flows through a monolayer filter that collects radon progeny in a thin layer at the surface of the filter. As the progeny decay, about half of the alpha particles are emitted into the volume above the filter and ionized in the surrounding air. Using an electric field, this ionization produced in the air above the filter can be collected and inputted into amplifiers which output a spectroscopy signal suitable for pulse height analyses. The measurement of radon and progeny using multi-wire air ionization chambers is an established technique; however, coupling the ionization chamber to the filtering system to allow simultaneous sampling and analysis is a new concept. One of the difficulties with measuring ionization using this technique is that the signals are very small and mechanical vibration of the wires causes electrical noise. However, in the method under investigation no wires will be used and the filter support assembly can be made very rigid, thus greatly reducing this problem. The spectral resolution will be poorer than for an ionization chamber alone, due to loss of alpha energy in the filter paper, but should be adequate to distinguish the various radon and radon progeny alpha signals.

  In essence, the instrument envisioned is a flow-through detector that can be made quite large. This permits the use of filters 6-inches and greater, which is necessary because typical monolayer filters (2 in-3 in) have a very high resistance to air flow. As for the Radgrabber, described in Summary No. 5.5, the sensitivity depends on the amount of air sampled per unit time and the large filter should allow large quantities of air to be sampled in a short time. This will permit measurements of very low radon progeny concentrations. Unlike the Radgrabber, which measures only charged progeny, this instrument will be able to measure both charged and neutral progeny. A number of potential problems need to be investigated prior to the design of a bench prototype sometime in 1995.

  Summary No. 3.9 also describes another new radon detector being developed in support of our Atmospheric Chemistry Program Research.

- LEA Instrument Development: The LEA aerosol measurement instrument package, described in last year's report, was completed and test flown in 1994 (see Summary No. 4.5). This instrument package was subsequently modified and installed on a tower at the ARM site to make baseline aerosol measurements (see summary No. 4.2). A report describing this instrument package will be published in 1995 (Albert and Leifer, in press).

- Four Port Parallel Impactor: The lightweight four-port parallel impactor described in last year's report was integrated into a complete instrument package, including: a step motor drive, four flow sensors, pump control, and a computer for data acquisition and sequencing. This instrument package will be used in the upcoming Gulfstream flights scheduled for the summer of 1995 (see Summary No. 5.5).

- Optical Particle Counter: An optical particle counter was integrated into an instrument package containing meteorological sensors and a computer. This instrument package was used at the ARM site and on the roof at EML to obtain particle size distributions (see Summary No. 4.5).

- ARM Site Sampling Stack & Manifold: The stack and manifold described in last year's report are ready for deployment, and installation is expected to take place in 1995. A detailed document, "statement of work", was prepared

and submitted to the ARM site Program Manager. This document, which includes many detailed drawings that had to be made, describes the procedures required to prepare the site and install the stack/manifold. A great deal of effort requiring both engineering and wiring continues in order to build a complete data system here at EML which duplicates the installation at the ARM site. It is expected that a contract for installing this system at the ARM Southern Great Plains site in Lamont, Oklahoma will be in place in the early fall of 1995, and the installation should begin shortly thereafter (see Summary No. 4.2).

- High Altitude Neutron Spectroscopy: A dual spectroscopy amplifier was designed and built in support of the Radiation Physics Division's high altitude neutron measurement program (see Summary No. 2.7). The design incorporated a new generation of high speed amplifiers and used computer aided design techniques for the required printed circuit boards. The entire system was successfully tested in a preliminary flight over Northern Canada.

- HPIC Systems: Three of EML's high pressure ionization chamber instruments described in last year's report were modified for use at the Princeton TFTR (see Summary 2.2).

- RAMP: A continuing effort is the maintenance of RAMP systems, including repair of units returned from the field and sending replacements. During 1994, a MCA was sent to Cape Point; transmitter, batteries and antenna cable to Pretoria; batteries and power supply to Marsh; a replacement UPS unit to Turkmenistan, and a system power supply to Perth. Custom-made test equipment with detailed instructions was shipped to Turkmenistan, which allowed the identification and solution of a difficult problem. Questions relating to RAMP operation are routinely answered by FAX, E-mail, letters and telephone.

  Additional RAMP systems were constructed using both NaI and HpGe detectors. A new RAMP unit (NaI/Modem) was installed at Eileson Air Force Base in October. During 1994, we also purchased and evaluated an Ortec Nomad, a Canberra Inspector and a Magnavox Satellite system. A "Call Ramp" program and later enhanced versions were extensively beta tested. Trips were made to PGT in Princeton, NJ, to inspect and perform acceptance tests on HpGe detectors purchased for RAMP sites.

  In the course of designing the automatic RAMP system described in Summary No. 1.5, a wood and cardboard mockup of two methods of implementing the automatic pick-and-place mechanism were constructed. This allowed a three dimensional visualization of the proposed methods, and enabled a quick decision on the method best suited for this application.

- Mercer cell: A new Mercer cell to measure the particle size distribution in the range of 0.5 to 10 nanometers was designed and built for the Radiation Physics Division (see Summary No. 3.13).

- Integrating bronchial dosimeter: A new integrating bronchial dosimeter was designed and construction initiated for the Radiation Physics Division (see last year's report).

As in previous years, a number of projects were carried out in 1994 to either improve or maintain the Laboratory's facilities. The Perimeter Security System for the 5th floor and lobby required regular maintenance, including work on door switches, replacement of a zone board and many indicator lamps. A system of monthly

inspections has been established, with resulting documentation. The smoke detection system required moving some detectors to accommodate contractors, as well as troubleshooting and repair to correct several zone problems. Additions were made to the system to include a hood and duct alarm notification in the lobby area.

A 60KVA UPS unit was installed adjacent to the Computer Room to maintain power to EML's VAX cluster during electrical transients and temporary shutdowns. Heat load was evaluated and an air conditioner was put in place, as well as ceiling vents to prevent battery gas buildup. An a.c. electrical distribution system, allowing two bypass methods and tying the UPS with the computer, was installed. The system is fully operational and is checked on a daily basis.

The EML machine shop was upgraded by installing automatic numerical control on a milling machine and installing a new miller/driller machine with digital readout. CAD facilities were upgraded with the installation of Autocad version 13.

Other examples of maintenance and/or improvement of Laboratory experimental equipment during 1994 include: design and construction of a new lead shield for the Analytical Chemistry Division germanium diode gamma-ray counting facility; support for the electron microscope facility; and installation of additional cables to extend the Laboratory's local area network (LAN). The Division also continued to maintain and support the EML Chester, NJ, regional baseline monitoring station.

Reference

Albert, B. and R. Leifer
"Electronic Design for the Light Experimental Aircraft Aerosol Package"
USDOE EML Report, in press

6.3 LABORATORY COMPUTER CENTER

Camille G. Marinetti, Robert Stocco and Jenny May

Facility Enhancements. The Laboratory's central computer cluster was upgraded with the replacement of two VAX-11/750's with a Digital Equipment Corporation VAX 6410, which has seven times the CPU speed of an 11/750. The amount of memory on both VAX's was increased to 64 Megabytes. New printers were added to the computer center during 1994. The DEC LP25 line printer was replaced with a DECLA424 multi-printer for wide carriage printing. Two Hewlett-Packard LaserJet 4P printers were added for network and VAX users. All of the shared Laboratory printers were moved out of the computer room for easier access by users and increased computer room security. An ENCAD Novajet color poster printer, a Seiko ColorPoint 2 dual dye-sublimation and thermal wax printer, and a Nikon LS3510 high resolution slide scanner were added to the graphics resources. The film recorder was upgraded to a Laser Graphics LFR Mark II. Twenty more users, four new PC software packages, and a PC network backup package were added to the PC LAN.

Computer Graphics. EML personnel can use the graphics facilities in Applied Mathematics to produce slides, overheads, posters and near photographic quality prints in-house. Users can create their own graphics on their IBM compatible or MacIntosh PCs, or give the Applied Mathematics staff the raw material and have the finished graphic presentation done for them. The graphics center printers and film recorder (for slides) are compatible with any Windows graphics software and most MacIntosh packages. Output from some DOS software can also be used. A flatbed scanner and a slide scanner allow direct input of graphics, photographs, and textual material. The NovaJet

poster printer can produce posters up to 2 feet by 3 feet.

DEC Pathworks LAN. The VAX cluster with the DEC VAX 6310 and 6410 computers supports over 80 user accounts via an ETHERNET LAN and a system of terminal servers throughout the Laboratory. PC users can communicate and share resources between the VAXs, IBM compatibles, and MacIntosh computers. Ethernet thin wire coax cables connect PCs to the Ethernet thick wire spine which runs around the Laboratory and also connects the terminal servers to the VAX.

The DEC Pathworks PC LAN allows PC users to share files, printers, and software between the network PCs and the VAX cluster. At this time, 65 users are connected to the PC LAN. PCs attached to the LAN are able to use VAX printers and print queues, and to use PC software installed on the VAX disk drives and available through the network. PC software currently installed on the LAN includes WordPerfect (DOS and Windows), Microsoft Word, PowerPoint, Excel, Access and FoxPro; Quattro Pro, Paradox for Windows, Word for Word, and Harvard Graphics for Windows. A LAN-based backup package was added to allow automated backups of PCs over the network during off-hours.

PC Repository. Used PC parts, peripherals, and programs have become a significant Laboratory resource. The PC repository is a central pool of used hardware and software. When employees contribute items which they no longer need, the item's condition and usefulness are assessed. If it is in working order and potentially valuable to another employee, it is stored in the repository. The items can then be redistributed where needed or used as replacement parts.

6.4 GC/MS DATA PROCESSING

Ada Kong and Yulin L. Tan

The Hewlett-Packard (HP) 5988 gas chromatography/mass spectrometry (GC/MS) system used for organic analyses is computerized. The software program, HP pascal ChemStation, controls the instrument operation, data acquisition, and data reduction. Although the data reduction can lead to final report generation, the reduction process is very limited and the reporting format restricted. For research, the need to transfer data files to a PC, where unlimited software can be used for various data processing and report generation, was apparent.

Since HP Pascal ChemStation does not allow direct file transfer to a PC, a platform using HP E2080A, LIF (logical interchange format) utilities for the PC has been established. The acquired data file is first reduced to an integrated file, then a report file. Through the report file the data can be copied onto a PC disk as an ASCII (text) file. Using LIF utilities in the PC, the ASCII file is converted into an MS-DOS text file. The converted file can then be processed by any PC application software.

We have processed our GC/MS data through these converted files with spreadsheet software for single run calculations, multi-run consolidations, statistical calculations, and report generations in both graphical and tabulated forms.

6.5 STABILITY OF DEUTERATED PAH

Yulin L. Tan and Ada Kong

In conducting polycyclic aromatic hydrocarbon (PAH) analyses by GC/MS, deuterated PAH isotopes are widely used as internal standards and calibration compounds to establish response factors for quantifications. We have found that some of the deuterated isotopes in a common hydrocarbon solvent, toluene, are not stable over a period of time. The degradation leads to changes in concentrations in standard solutions and response factors in calibrations, and, consequently, casts doubt upon the reliability of quantifications.

The most likely causes of the degradation of deuterated isotopes in a hydrocarbon solvent is the exchange of deuterium atoms on the isotopes with the surrounding hydrogens on the solvent molecules. To confirm this speculation, PAH solutions in both deuterated and native toluene are being analyzed.

Series of calibration solutions containing various deuterated and native PAH, and standard solutions containing either deuterated or native PAH are being made in both deuterated and native toluene. These solutions will be periodically analyzed over a year period by GC/MS under selected ion monitoring mode. Standard solutions of native PAH will be monitored at molecular ion (M), M+1, and M+2, standard solutions of deuterated PAH at M, M-1, M-2, and calibration solutions at M.

The results from this study should indicate, within a year, the stability of various deuterated and native PAH in deuterated and native toluene, and the consistency of response factors generated from the calibration solutions of these PAH in deuterated and native toluene. These results will provide guidance in making stable deuterated PAH solutions as internal standards and calibration solutions yielding consistent response factors.

6.6 ACTINIDE METROLOGY: CHROMATOGRAPHIC RESINS FOR THE ISOLATION OF ACTINIDES

Anna Berne

A new procedure for americium determination on Quality Assessment Program (see Summary No.7.4) filter samples has been validated by other analysts at EML (see Table 6.1). As can be seen from the table, no significant bias was observed. This procedure was previously tested and the results were compared with those obtained using the standard EML procedure, Am-01 (see last year's Annual Report). Both sets of analyses were performed by the same analyst.

The procedure is applied to the preparation, separation, and analysis of spiked water and air filter samples that contain americium but not lanthanides based upon the use of Eichrom's TRU Resin. Combined with EML's Procedure Pu-11, this procedure allows for the sequential determination of plutonium and americium.

--- Eichrom's TEVA resin column has been used at EML successfully instead of a thiocuanate column to remove lanthanides from soil samples that are being analyzed for americium. The chemical recoveries of americium in soils have been increased from an average of 30% to an average of 70%, and the time of analysis has been shortened by 4 working days. The use of this resin will be included in the Am-01 Procedure (Berne and Greenlaw, in press).

--- A method utilizing two different extraction materials (Eichrom's TRU and TEVA Resins) was used as an effective and quick solution to a problem encountered during analyses of Mixed

TABLE 6.1

COMPARISON OF TWO METHODS USING EML PROCEDURE
Am-01 AS THE STANDARD*

	Sample ID	New Am. Proc.	Am-01 Proc.	% difference
Water (Bq L^{-1}):	9409	1.01 ± 0.06	1.01 ± 0.06	-3
	9503	1.32 ± 0.11	1.35 ± 0.01	-2
Air (mBq):	9409	214 ± 9	214 ± 9	+2

* Error is the standard deviation of the mean of 3 replicates.

Analyte Performance Evaluation Program (MAPEP) soil samples. The alpha spectrometry analysis of the microprecipitated americium fraction showed heavy contamination with thorium which had to be removed. The results of this emergency response were presented at the 40th Annual Conference on Bioassay, Analytical and Environmental Radiochemistry at the Eichrom Industries, Inc. Workshop on November 14, 1994 in Cincinnati, Ohio.

--- The preliminary results of a new procedure for the determination of plutonium and americium in soil aliquots, 5-50 g in size, will be presented at the 41st Annual Bioassay Conference.

Reference

Berne, A. and P. Greenlaw
in: "EML Procedures Manual"
USDOE Report HASL-300, 28th Ed.,
Vol. 1, in press

6.7 GAMMA-RAY SPECTROMETRY

Colin Sanderson, William Rivera and
Karin M. Decker

Low level-environmental gamma-ray spectrometry is often complicated by a wide variety of sample matrices, different types of detectors and numerous counting geometries. Environmental samples collected for gamma-ray spectrometry usually consist of soil, vegetation, tissue, water or air particulates. These samples may range in size from a few grams or cubic centimeters to many kilograms or liters.

Gamma-ray spectrometry at EML is performed with eight high resolution germanium detectors in five different configurations. These consist of three 1.5 cm diameter wells, a 1.5 cm well in a segmented germanium detector with a sodium iodide Compton suppression shield, three n-type low energy coaxial, and a 2000 square

centimeter planar detector. Gross gamma-ray screening is obtained from the events detected with the coaxial germanium systems. Four different counting geometries are routinely used for quantitative analysis. These are 600 cm^{-3} Marinelli (reentrant) beakers, 90 cm^{-3} aluminum cans, 45 cm^{-3} plastic planchetts, and 1 to 2 cm^{-3} samples in plastic test tubes for well counting.

During the past year, 1329 filter samples from the Environmental Studies Division's Surface Air Program were routinely analyzed with germanium detectors for ^{7}Be, ^{95}Zr, ^{137}Cs, ^{144}Ce and ^{210}Pb. In addition, over 1380 Quality Assurance Program (QAP) intercomparison, and miscellaneous samples were analyzed for a variety of gamma-ray emitters using these systems.

6.8 SEMI-EMPIRICAL ANGULAR RESPONSE FOR GERMANIUM DETECTORS

Peter Shebell and Marcel Reginatto

The calibration of a germanium (Ge) detector for *in situ* spectrometry involves the determination of its angular response. The current EML procedure uses a point source at a fixed distance of at least 1 meter and measures the full absorption peak count rates at 15° intervals between 0° and 90°. The data is then fit to a polynomial. This procedure is simple but tedious. Furthermore, errors can result when poor statistics for weak gamma emitters cause fluctuations in the data which are incorporated into the polynomial fit.

A general semi-empirical procedure for determining the angular response for coaxial Ge detectors is being developed. The approach considers the surface area of the detector that intersects the photon beam, as well as the absorbing properties of materials comprising the detector. The procedure will involve a fit to a single parametrized function of the form:

$$R(\theta,E) = A(\theta,E) + B(\theta,E)\cos\theta + C(\theta,E)\sin\theta$$

where R is the detector's relative angular response, and A, B, and C are the coefficients that contain the parameters. The function will cover a full range of coaxial Ge detectors and photon energies. This new procedure will reduce the number of measurements required for calibration and improve the accuracy of the detector's angular response.

Preliminary results for a p-type closed end coaxial detector with a relative efficiency of 41.8%, and a crystal length-to-diameter ratio of 1.32, indicate that a function involving a single free parameter will agree with measured data to within 2%.

6.9 *IN SITU* GAMMA-RAY SPECTROMETRY

Kevin Miller, Peter Shebell and Gladys Klemic

Gamma-ray spectrometry is used routinely at EML to both identify and quantify radionuclides directly in the field. This provides measurement results immediately without recourse to sampling. The needs within DOE with regard to environmental restoration as well as those for other government agencies and international groups that assess contamination have expanded greatly in recent years. EML has responded to several dozen inquiries in the past 4 years from investigators seeking information and guidance in the application of this technique. To further enhance the adaption of this technology, we have organized and conducted a short course on *in situ* gamma-ray spectrometry for site assessment in

3 of the past 4 years for the IEEE Nuclear Science Symposium. At the 1994 meeting in Norfolk, VA, 24 individuals from various private companies and government and international organizations took this course. Aside from the theory and applications presented by EML personnel, representatives from the Pacific Northwest Laboratories, the TMC Company and the Yankee Atomic Electric Company presented applications.

In 1994, EML was under contract to the Nuclear Regulatory Commission to develop applications of *in situ* spectrometry for decommissioning surveys (see Summary 6.10). We also began to negotiate funding to provide consultation to contractors at the DOE Weldon Spring Remedial Action Site for characterization surveys and the DOE Nevada Operations Office for emergency measurements under the auspices of the Federal Radiation Monitoring and Assessment Center.

Since 1991, an EML representative has served on a report committee devoted to *in situ* spectrometry for the International Commission on Radiation Units and Measurements (ICRU). This document will provide standard guidance on the technique with some degree of international consensus. Final editing of this report was completed in 1994 and publication is expected in 1995 (ICRU, in press).

Extension of the technique of in situ spectrometry can be made to the x-ray region as well. For instance, characteristic x-rays from transuranics can be measured for quantifying the levels of a Pu isotopic mix in surface soil. In 1994, a presentation on EML's use of large area proportional counters for this purpose was made by invitation at a special session of the American Nuclear Society Meeting devoted to "The Use of Nuclear Instrumentation in Environmental Monitoring and Characterization". Our results indicate that the lower limit of detection (LLD) using this counter and a ten minute count would be approximately 300 Bq kg^{-1} (8 pCi g^{-1}) for a typical weapons grade Pu mixture in surface soil. For a freshly deposited source on top of the ground, the LLD would be about 200 Bq m^{-2} (5 nCi m^{-2}). These values are several times better than those achieved with traditional measurements using thin scintillators.

References

International Commission on Radiations Units and Measurements
"Gamma-Ray Spectrometry in the Environment"
ICRU Report 53 (in press)

Miller, K. M.
"A Large Area Proportional Counter for In Situ Transuranic Measurements"
Trans. American Nuclear Society, 70, 47-48 (1994)

6.10 CONSULTATION TO THE NRC

Kevin M. Miller, Carl V. Gogolak,
Peter Shebell and Gladys Klemic

EML entered into contract with the Nuclear Regulatory Commission (NRC) in 1993 to provide consultation on the development of new decommissioning criteria. A proposed rule on the radiological criteria was published in the Federal Register in August of 1994. This rule has as its basis a concept of "indistinguishable from background" for residual radionuclides in order for a site to be released for unrestricted use. It is an important, precedent setting rule that could have a major impact on the course of environmental cleanups at NRC licensed facilities as well as those associated with the DOE and the Department of Defense (DOD). To support this rulemaking, EML developed sections of the appendix to the draft generic Environmental Impact Statement. Our work dealt with the variability and measurement of background radiation. This information

was published this past year as a separate report to serve as a primer for interested parties (Huffert et al., 1994).

In 1994, our work for the NRC expanded to include the development of both a measurement and statistical testing methodology that would be used for performing radiological surveys for decommissioning. This work serves to provide a technical basis to support and implement the proposed cleanup criteria. The proposed criteria for unrestricted use are twofold: 1) 0.15 mSv (15 mrem) per year maximum total effective dose equivalent (TEDE), and 2) an ALARA (as low as reasonably achievable) goal extending down to 0.03 mSv (3 mrem) per year for radionuclides distinguishable from background.

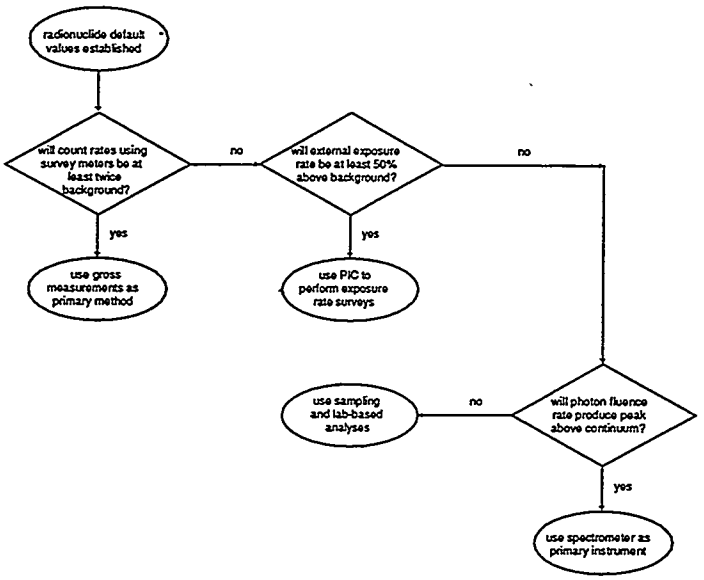

Figure 6.1 Flow diagram for determining the need for relying on more sophisticated measurements for decommissioning surveys.

The proposed cleanup levels represent relatively small increments above a background which itself has variations that can be as large as the increments. Presently used statistical methodology which rely on the Student's t-test can result in inefficient survey designs for determining compliance with the new decommissioning criteria. In some cases, this could lead to an unacceptably high error rate in decommissioning decisions. We have adapted and are recommending alternative methods based on the use of nonparametric statistics. This approach has the potential to reduce the number of samples needed to provide a defensible basis for objectively planning, executing and interpreting final status surveys. Specifically, the Wilcoxin Rank Sum Test, the Quantile test, and simple "hot spot" test can be applied to determine if the site meets the release criteria. A draft report on survey planning design incorporating this methodology was prepared for comment in 1994 (Gogolak, in press).

As a companion document to the statistical methodology eport, a draft report on the measurement methodology suggested by EML for decommissioning surveys was prepared during 1994 (Miller, in press). Specific instruments, measurement modes and types are being recommended in keeping with the calculated default concentrations for individual radionuclides that correspond to the 0.15/0.03 mSv level. Separate default values exist for the different scenarios (building occupancy, residential, etc.) that might occur following release of a decommissioned site. In some cases, the radionuclide concentrations would be difficult to measure using standard survey meter techniques. For this reason, nuclide specific measurements would be the best approach. This would involve, for instance, the use of *in situ* gamma-ray spectrometry. Using the data quality objective (DQO) approach, a cost effective mix of scanning, dose

rate, spectrometer and sampling measurements would be formulated. As an example of the type of guidance that could be used, Figure 6.1 shows a decision making flow chart.

Our work in this area will continue with the training of NRC staff and their contractors, and the demonstration of the measurement techniques at a number of licensed facilities that are undergoing decommissioning. In addition, at the request of the NRC, we are participating as representatives on the Multi-Agency Radiation Site Investigation Manual (MARSIM) Committee. This committee will attempt to establish a uniform approach to surveying and testing for potentially contaminated sites around the country that are under the jurisdiction of various agencies.

References

Gogolak, C. V.
"Non-Parametric Statistical Methodology for the Design and Analysis of Final Status Decommissioning Surveys"
USNRC Report NUREG-1505 (draft), in press

Huffert, A. M., R. A. Meck, and K. M. Miller
"Background as a Residual Radioactivity Criterion for Decommissioning"
USNRC Report NUREG-1501, in press

Miller, K. M.
"Proposed Methodologies for Measuring Low Levels of Residual Radioactivity for Decommissioning"
USNRC Report NUREG-1506 (draft), in press

6.11 ENVIRONMENTAL RADIATION MEASUREMENTS AT THE FORMER SOVIET UNION'S SEMIPALATINSK NUCLEAR TEST SITE

Adam R. Hutter and Peter Shebell

Two EML scientists served as scientific experts at the invitation of the IAEA to participate in its mission to Kazakhstan during July 1994. The former Soviet Union's largest nuclear test site was located near Semipalatinsk, Kazakhstan, and following Kazakhstan's independence, the IAEA committed to studying the environmental contamination and the resulting risk to the population from radiation exposure from the nearly 500 tests (including underground, atmospheric and surface detonations) performed at the site between 1949 and 1989. Scientists from France, the United Kingdom, Russia, and Austria comprised the remainder of the 11 member team.

The purpose of the mission was to perform a radiological preassessment of the former Soviet nuclear test site at Semipalatinsk and surrounding villages to determine if a full assessment would be warranted. During the 2-week expedition to Semipalatinsk and surrounding villages, the EML scientists provided external gamma dose rate measurements, *in-situ* gamma spectral analyses, and they collected soil samples for a radiological preassessment of the current conditions of the radiation dose to the local population. The team surveyed areas within the uncontrolled test site,

the surrounding villages of Kainar, Sarzhal, Dolon, Dolok and Akzhar, as well as several farms and grazing pastures (see Figure 6.2 for site locations). Within the test site, the team performed measurements to gather radiological data as well as to corroborate previous measurements at the historical "ground zero" and surrounding areas, two "excavation lakes" created by underground testings, and two "hot spots" that were determined to be in the path of the plume following weapons tests, as denoted from maps supplied by the Russian government, of previously performed aerial gamma surveys. Residences and public areas in the villages near the test site were extensively surveyed. It was important to consider all exposure pathways, including dietary samples. The diet of the local population consists mainly of the meat and milk from sheep, horses, goats and cows, and is nearly devoid of fruit and vegetables. Therefore, biological (i.e., bone, meat, offal, milk), soil, water, and vegetation samples were collected. Also considered as an ingestion pathway was the inhalation of resuspended material.

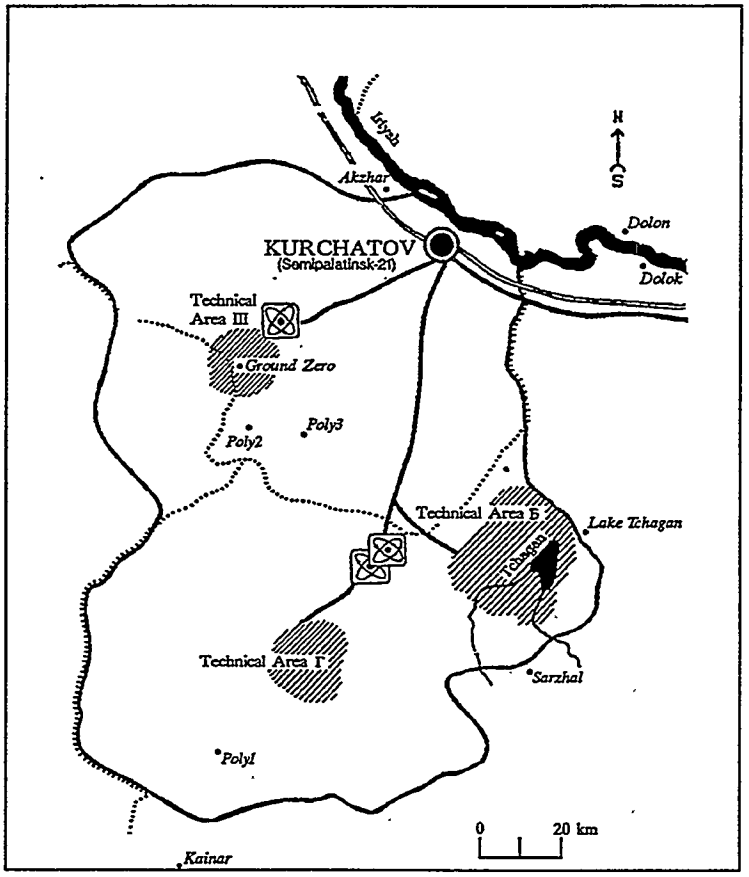

Figure 6.2 Map of Semipalatinsk nuclear test site showing locations where radiation measurements were obtained as part of a radiological preassessment to determine the current radiation risk to the population.

In all, the EML scientists collected 19 *in-situ* gamma-ray spectra taken with a germanium detector, ~200 dose rate readings measured with pressurized ionization chambers, and 12 soil profiles sampled with 8.9 cm diameter corers.

Many of the sites where the team performed measurements were found to be near "background" levels. However, human exposures could be minimized if control measures were put in place to inhibit access to certain areas by nomads, settlers, and grazing animals.

A report will be forwarded to the IAEA as soon as the soil samples have been analyzed for inventory estimates of ^{137}Cs.

6.12 FALLOUT IN NORTH DAKOTA FROM NEVADA WEAPONS TESTING

Harold L. Beck *

On January 16, 1994, Senator Byron Dorgan, North Dakota, wrote the Secretary of Energy requesting all available information on fallout from Nevada weapons tests and other atmospheric tests in North Dakota. The Senator was concerned as a result of news articles suggesting fallout levels in North Dakota may have been higher than originally reported and that all the original data may not have been released. In cooperation with the Nevada Operations Office, EML prepared a summary of available information on fallout in North Dakota for inclusion in the response to the Senator. Additional material and specific responses to the Senator's concerns were addressed by Nevada Operative Office. The Nevada Operations Office also provided voluminous reference material to the North Dakota Health Department from the DOE fallout archives in Las Vegas.

Using this material the North Dakota Health Department prepared a draft report for the Senator. This report suggests that parts of North Dakota may have received an unusually high deposition of ^{90}Sr from both the Nevada Test Site (NTS) and global fallout and that some residents may have contracted certain types of cancer from exposure to fallout and suggests the need for additional studies. On December 16, 1994, a briefing was held for the Senator in his office to discuss this report and his concerns. Harold Beck, EML Acting Deputy Director, participated in this briefing along with representatives from the National Cancer Institute (NCI), other DOE units, and the North Dakota Health Department. EML was invited to participate based on the major role it played in all recent reassessments of NTS fallout and the fact that most of the relevant data was collected by EML. The NCI representatives briefed the Senator on the various epidemiological studies that have been carried out to assess the impact of NTS and global fallout. They also discussed radiation exposure risk factors and attempted to put the fallout doses in perspective with other risks and with natural background radiation exposure.

EML's analysis of available data does not indicate fallout levels and related radiation exposures in North Dakota were unusual compared to other areas of the U.S. However, the Senator has requested that the DOE and the National Institute of Health, in cooperation with the North Dakota Health Department, institute further investigations of fallout levels and related health effects in the Mountain Plains States. If additional assessments are carried out, it is expected that EML scientists will be asked to participate or consult in that effort.

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* In consultation with Edward P. Hardy, Jr.

6.13 ^{85}KR AND ^{129}I IN THE ENVIRONMENT

Harold L. Beck

At the request of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) secretariat, EML conducted a literature review to gather data on the sources, levels, and resultant population doses for ^{85}Kr and ^{129}I in the biosphere. Of particular interest to UNSCEAR was the availability of actual monitoring data at various sites around the world that would allow current and future geographical variations in concentration to be determined. It was found that ^{85}Kr had been measured routinely at a number of sites over long intervals. However, only sparse information is available for ^{129}I, mostly downwind from nuclear facilities. The available ^{85}Kr data has been used extensively to test global atmospheric transport models and a number of investigators have published calculations of concentrations as a function of latitude and longitude.

The information gathered was provided to the UNSCEAR secretariat and will be used as source material for the preparation of the next version of the UNSCEAR report on sources and effects of ionizing radiation.

7

Environmental Management Programs

7.1 OVERVIEW

Catherine S. Klusek

In its five year plan, the Office of Environmental Management (EM) specifies that policies be instituted to establish, assess and maintain high quality analytical laboratories to provide the quality data needed to fulfill the needs and requirements for the operation of restoration and waste management at DOE sites. The Office of Compliance and Site Coordination, through its Analytical Services Division (EM-263), has designed and is implementing an Analytical Services Program (ASP) to address these issues. Under the direction of EM-263, EML is currently involved in three components of the Quality Assurance (QA) Program elements of the ASP: the QA Guidance and supporting documents, the Performance Evaluation Programs, and the Assessment Program. The objective of the QA element of the ASP is to develop, integrate and implement a comprehensive QA program, including an assessment function that addresses the full scope of a laboratory's performance from sampling through analysis to data evaluation.

In addition to EML's core effort in EM-263's QA programs, described above, other project are undertaken on a fiscal year basis depending on the needs of headquarters and staff availability. These activities are of two types: (1) direct support to EM-263 in addressing issues relevant to DOE's environmental needs and planning efforts through participation in working groups, presentations and technical reviews; and (2) technical activities to continually improve the analytical capability of EML and DOE support contractors, and to introduce improvements or additional samples/ analytes to the ongoing performance evaluation programs. For example, EML participates on the EM Environmental Sampling and Analysis Coordinating Committee (ESACC), which is a forum for issues of interest to EM program managers, is a member of the review team for the DOE Methods Compendium, and pursued several areas of analytical development utilizing liquid scintillation techniques. Other activities which support EML's performance evaluation programs include participation in round robins sponsored by standards setting organizations (e.g., ASTM and IAEA).

Support for the Office of Technology Development (EM-50) continued through the efforts of EML's Technical Program Manager. Several additional technical activities, related to the former Soviet Union and baseline monitoring techniques, were also pursued which will have direct applications to DOE remediation efforts.

EM-263 QA Program Support

7.2 EM-263 QA GUIDANCE DOCUMENTS

Michael Johnson, Catherine S. Klusek and Hemant Pandya

Seven documents introducing EM-263's Quality Assurance Program elements were published (DOE, 1994a-f). These guidance documents address the requirements of DOE Order 5700.6C Quality Assurance for EM environmental sampling and analysis activities. EML staff participated in the Working Groups which drafted the documents. The topic areas include:

a) QA guidance pertaining to the design and implementation of sampling and laboratory procedures and processes for collecting EM environmental data;

b) A description of the Integrated Performance Evaluation Program (IPEP), which is designed to integrate information from existing performance evaluation (PE) programs and provide information about the quality of radiological, mixed waste and hazardous environmental sample analyses provided by all laboratories supporting EM programs.

c) Performance objectives and assessment criteria for QA management assessments; and

d) Assessment standards that can be used to conduct a performance appraisal of an organization's or projects's ability to meet quality goals for sampling, field measurements and analytical activities.

In 1994 additional draft documents were developed which address the following topic areas (DOE in press):

a) Performance objective and assessment criteria for assessment of sampling activities, as well as revisions to the QA management assessments document in this area; and

b) QA guidance pertaining to the design and implementation of field measurement procedures and processes for collecting EM environmental data.

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Draft document, in press

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Draft document, in press

7.3 THE INTEGRATED PERFORMANCE EVALUATION PROGRAM (IPEP)

Catherine S. Klusek

EML is currently participating in the IPEP Working Group with Argonne National Laboratory-East (ANL) and DOE's Radiological and Environmental Sciences Laboratory (RESL) in the development of EM-263's IPEP. The program will provide information on the quality of radiological, hazardous, and mixed analyte analyses performed under contract to DOE by acting as a clearing house for performance evaluation results from programs sponsored by DOE and the Environmental Protection Agency (EPA). Through the Working Group, plans were developed to pilot the IPEP database in conjunction with a selected DOE Office of Sample Management. This approach, which will be initiated with EM-263's Resource Management Group, is being carried out by ANL.

EML's specific area of responsibility will be as sponsor of the DOE Quality Assessment Program (QAP). The DOE QAP will serve as a radiological performance evaluation (PE) component of the IPEP. The QAP is discussed in Summaries Nos. 7.4 - 7.7.

EML will also participate in the DOE Mixed Analyte Performance Evaluation Program (MAPEP) sponsored by RESL. This program will provide information on the quality of data provided for mixed waste analyses performed under contract to DOE. EML is one of three first-tier laboratories who will verify the analyte concentrations in the prepared materials prior to distribution. Activities in MAPEP for this year are detailed in Summary No. 7.8.

7.4 QUALITY ASSESSMENT PROGRAM (QAP)

Colin G. Sanderson, Catherine S. Klusek, Pamela Greenlaw, Isabel Fisenne, Vivian Pan, Anna Berne, Steve Minick, JoAnn Morris, Pamela Perry, William Rivera, Salvatore Scarpitta, Marie Lawrence and Richard Godwin

The EML QAP is designed to test the quality of the environmental radiological measurements reported by contractor laboratories to DOE Offices of Environment, Safety and Health (EH) and Environmental Management (EM). Under QAP, natural matrix or synthetic environmental

samples which have been prepared and thoroughly analyzed at EML for as many as 20 radionuclides are distributed semiannually to the contractors and other participating laboratories. Each participant receives 8-10 samples consisting of soil, vegetation, tissue, air filters and water samples spiked with radionuclides. Most of the soil and vegetation samples have been collected from locations where the radioactive concentrations in these matrices are known to be higher than average background values.

During 1994, QAP samples were distributed on March 1 (QAP40) and September 1 (QAP41), and two EML reports were issued. EML-559 (Sanderson and Klusek, 1994a) covered the participants results for the QAP39 sample distribution. EML-551 (Sanderson and Klusek, 1994b) covered the participants results for the QAP40 sample distribution.

During this past year, 129 laboratories received QAP samples. This was an increase of 20 over 1993, an increase of 35 over 1992 and an increase of 55 over 1991. Ninety-eight laboratories reported data for the QAP39 sample distribution and 101 reported data for QAP40.

The total number of analyses performed by all participants in 1994 (QAP39 and QAP40) was 4675, about 7% more than was reported in 1993. In general, the 1994 data reported continues to be satisfactory. For 1994, 75% of the reported results were within 20% of the EML value, 8% differed from the EML value by more than 50%. In 1993 about 7% of the results differed from the EML value by more than 50%.

Acknowledgement -- The following support personnel contributed significantly to the success of this program: Sylvia Hulse, Andrea Delgado, Kevin J. Clancy, Arnold Boyd, William Jackson, Camille Marinetti, Herbert W. Feely, Marcel Reginatto, Brenda O. Jones and Nancy Chieco.

References

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USDOE Report EML-561, July (1994b)

7.5 QAP OPERATIONAL CRITERIA AND PERFORMANCE EVALUATION

Vivian Pan

Operational criteria and control limits for QAP analytical performance have been derived from historical QAP data. Beginning with QAP distribution 9409 (QAP 41), performance of individual reported data will be evaluated based on these criteria. The performance criteria are determined from observed analytical capabilities over a 10 year history of the program. Individual analyte/matrix control limits are established from percentile distributions of cumulative historical values which are normalized to EML's values. Three categories of performance have been established: acceptable, acceptable with warning, and not acceptable. The operational control limits and performance criteria are further discussed in Pan (in press).

The performance evaluation was first piloted with QAP40 (9403) data to ascertain the validity of the control limits. Results of the evaluation show that performance proportions in the three categories for QAP40 data are consistent with those of previous QAP intercomparisons which used ± 20% and ± 50% as control limits.

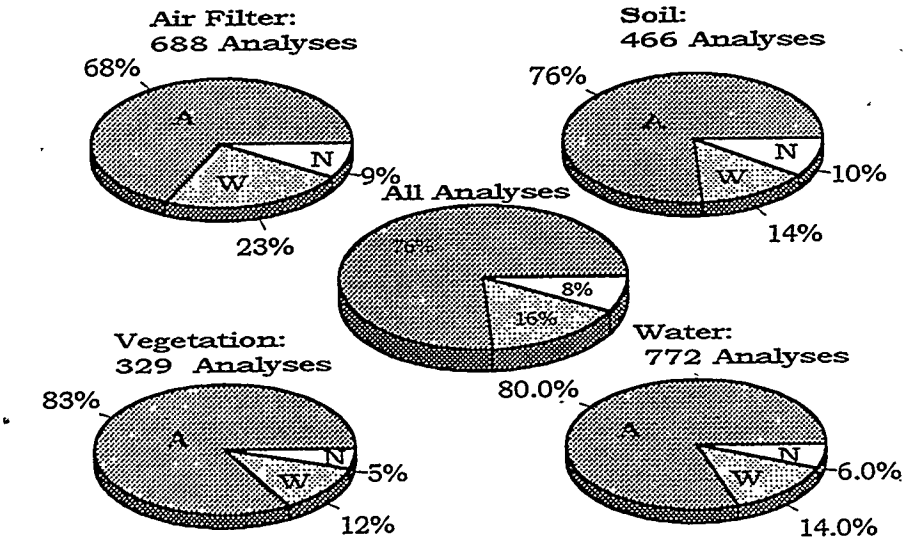

Figure 7.1 QAP 40 Summary of evaluations for 2255 reported analyses.

Figure 7.1 displays the proportions of performance for 2255 reported analyses per matrices of air filter, soil, vegetation and water, and in total. The first treatment of this evaluation system will be administered in the QAP41 (9409) distribution.

References

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"Analysis of EML QAP Data from 1982 -1992: Determination of Operational Criteria and Control Limits for Performance Evaluation Purposes"
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7.6 GROSS ALPHA AND BETA DETERMINATIONS IN QAP WATER AND AIR FILTER SAMPLES

Salvatore C. Scarpitta*

One component of the DOE EM-563 Quality Assurance Program is participation in the EML Quality Assessment Program (QAP) by contractor laboratories conducting work for the DOE. The EML QAP is mandated by DOE Order 5400.1, Ch 4.10.c and EM Secretarial memorandum. The EML QAP is a DOE complex-wide effort that provides analytical evaluation needed to address data useability for environmental restoration and waste management problems and requirements

*Project was carried out in conjunction with Tser-Min Chang of the Institute of Nuclear Energy Research, Republic of China

(see Summary No. 7.4). The 17 year old EML-QAP will be expanded to incorporate additional water and air filter samples for gross alpha/beta analysis. These samples will be distributed concurrently with the routine semiannual QAP samples. The first distribution of the samples for gross alpha/beta analysis will be in March 1995 (QAP42).

Instrumentation. EML has acquired a Tennelec/Nucleus Model LB4100W Windowless Gas-Flow Low-level Alpha/Beta Counting System (Oxford Instrument Co.). The efficiency of the system is nuclide specific, ranging from 30 to 60%, whereas the background counts per unit time is essentially an instrument specific parameter.

The 2" diameter detectors have a measured alpha background of 0.05 counts m^{-1} and beta background of 1.0 counts m^{-1}. The instrument has a lower limit of detection (LLD) of 37 mBq (2.2 dpm) for a 60-min count at the 95% confidence level with a 30% detection efficiency, whereas with a background of 0.05 counts m^{-1}, it will have an LLD of 7.4 mBq (0.4 dpm) for the same count time and detection efficiency (NCRP, 1978). The gridded proportional detectors reportedly eliminate "cross-talk" between the alpha and beta channels, allowing for simultaneous detection of gross alpha and gross beta counts. The basic unit consists of two drawers that each contain four independent detectors. Each drawer can be configured with four 1" or 2" detectors with individual guard detectors per drawer. Point sources of ^{90}Sr ($t_{1/2}$ = 27.7 y) and ^{210}Po ($t_{1/2}$ = 138.4 d) are available for both plateau voltage and efficiency calibrations. All data are processed with a 486DX (33 MHz) PC computer that operates in DOS and Windows and utilizes commercially available Data Analysis Spreadsheet software (that is, Excel, Quatro, Minitab) .

QAP Samples. Samples for gross alpha/beta counting to be distributed to DOE/QAP participating laboratories will consist of spiked water and styrofoam simulated "filter" samples. Master solutions of known activity are prepared for both water and filters using NIST traceable radionuclides calibrated at EML using a 2-π geometry detecting system.

The final 1 L spiked water master solution will typically contain <200 dpm mL^{-1} of an alpha/beta mix of either ^{90}Sr and $^{238,239}Pu$, ^{241}Am, or natural U. The 4 mL gross alpha/beta water samples (GW) are dispensed into precleaned glass vials with leak-proof Teflon sealed caps for the semi-annual QAP distribution.

Approximately 200, 1-7/8" diameter styrofoam discs are spiked using the air filter master solution. The final filter master solution would typically contain <2000 dpm mL^{-1} of an alpha/beta mix of either ^{90}Sr and $^{238,239}Pu$, ^{241}Am, or natural U. The actual spiking of the foam filters is performed using a mechanical semi-automated pipetter developed at EML, capable of delivering predetermined microliter amounts of tracer solution onto the filter surface. The pipetting unit is currently programmed to deliver 12 aliquots of 10 μL each onto a 2" diameter disc in a fixed

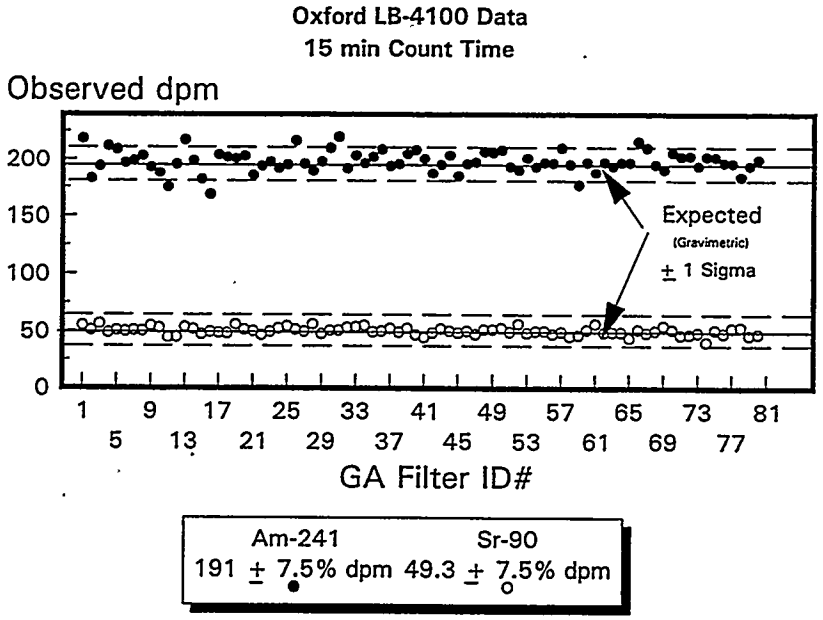

Figure 7.2 Summary of analysis of 80 GA filters.

geometric pattern and is accurate to within 8% of the preset amount. Each filter is individually counted for gross alpha/beta activity for 15 min in the Oxford LB4100W system to establish sample homogeneity, determine outliers, and establish the "EML Measured Value" (See Figure 7.2).

Quality Control. EML will participate in the USEPA Performance Evaluation Studies Program which is a QA program administered by the Nuclear Radiation Assessment Division of the USEPA at the Environmental Monitoring Systems Laboratory, Las Vegas (EMSL-LV), NV. Currently, the USEPA (Dilbeck and Honsa, 1980) provides 1 L water samples containing a mixture of radionuclides preserved with 0.5N HCl. These samples are distributed semi-annually and consist of two samples per shipment. Sample (A) contains ^{226}Ra, ^{228}Ra and natural U which are analyzed for gross alpha activity. Sample (B) contains ^{89}Sr, ^{90}Sr, ^{60}Co, ^{134}Ce and ^{137}Cs (<7.4 Bq L^{-1}) which are analyzed for gross beta activity. Foam filters of 2" diameter containing ^{241}Am, ^{90}Sr or ^{137}Cs (<7.4 Bq $filter^{-1}$) are also distributed annually by the EMSL-LV. These filters will be analyzed at EML for gross alpha and beta activities as part of EML's internal QA program for the QAP gross alpha/beta measurements (Goheen et al., 1994).

In addition, all EML QC data, which includes background data and calibration standards, will be maintained as part of the software package of the Oxford Alpha/Beta detection system. The data will be available for easy reference or inspection and production of QC charts. During gross alpha/beta filter and water sample counting, blanks are interspersed between the actual samples to determine if detector contamination is occurring.

Data Validation. A commercially available Packard Tri-Carb 2250CA liquid scintillation counter will be utilized to validate the activity concentrations (alpha and beta) of the master solutions used to prepare the QAP gross alpha/beta water (GW) and filter (GA) samples. The counting technique applies to clear liquid samples, and the method can be completed in 1-2 hr once the appropriate quench efficiency curves have been established. The liquid scintilation counting system at EML is a multisample, automatic system which can detect both alpha and beta particles. The system can be operated in either the "dual isotope" or "full spectrum analysis" (FSA) mode to discriminate between alpha and beta particles.

Typically, ^{90}Y/Sr and ^{241}Am can be used in this system in activity ratios ranging from 1:1 to about 1:4. The activity concentrations in the GW master solution will typically range from 10-100 dpm mL^{-1} ^{90}Sr and 50-200 dpm mL^{-1} of ^{241}Am, whereas the activity concentrations in the filter solution (GA) will range from 100-500 dpm mL^{-1} ^{90}Sr and 500-2000 dpm mL^{-1} of ^{241}Am.

Figure 7.3 shows the results obtained using FSA/liquid scintilation, dual dpm and Cerenkov counting to verify the activity concentrations of either ^{241}Am or ^{90}Sr in the GA and GW master solutions. Analytical procedures developed at EML for ^{241}Am and ^{90}Sr analyses (Am-02, Berne, in press; and Sr-03, Morris, in press) were modified to independently validate the activity concentrations of both GW and GA master available Eichrom extraction chromatographic resins (Eichrom Industries, Inc., Darien, IL) solutions. The method involves separating the ^{241}Am from the ^{90}Y/Sr using commercially specific for either ^{90}Sr (Sr-Spec) or ^{241}Am (Tru-Spec). Strontium-90 is quantified by Cerenkov counting ^{90}Y during build-up in the Sr fraction. Americium-241 is quantified by either liquid scintillation analysis counting or solid-state alpha spectrometry, with ^{241}Am co-precipitated with NdF_3.

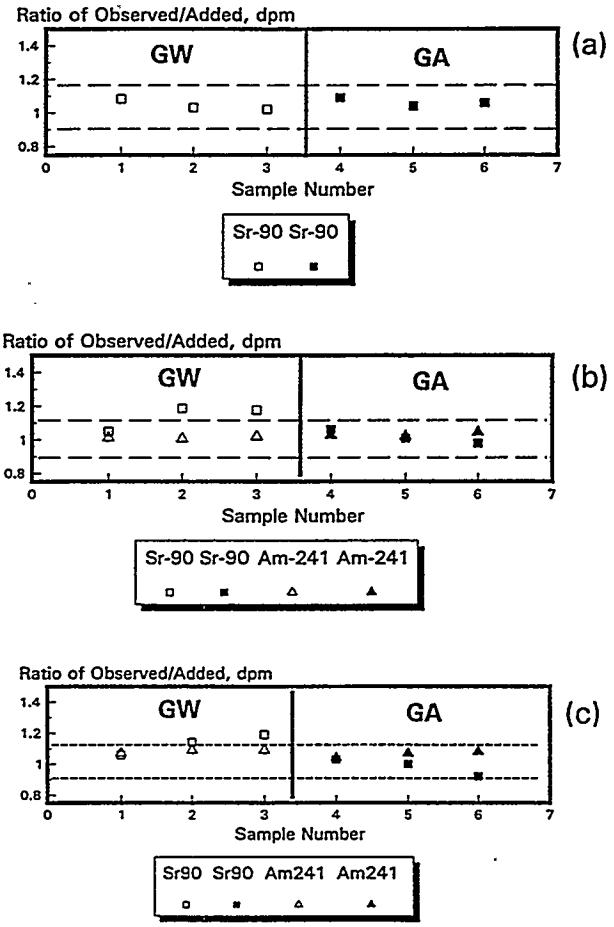

Figure 7.3 Verification of activity concentration in GA and GW master solution.

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7.7 GAMMA SPECTROMETRY DATA VALIDATION PROGRAM

Karin M. Decker and Colin G. Sanderson

EML will implement a Gamma Spectrometry Data Validation Program in 1995 in support of the DOE Office of Environmental Management (EM). The goal of this program is to assess the capability of DOE laboratories and DOE contractors in performing routine gamma spectra analyses required for EM projects, site evaluations and other DOE programs. Data reduction of gamma spectra are normally performed with computer codes supplied by commercial manufacturers or are developed "in house". Evaluation of some of the commercially available software was performed at EML in 1987 (Sanderson, 1988), and again in 1991 (Decker and Sanderson, 1992). The first study indicated there were substantial differences in the ability of the programs to detect small peaks and deconvolute overlapping peaks. The second evaluation showed most of the programs had fairly good results in peak detection and deconvolution, but the analysis of a complex spectrum still gave spurious results. Many of the available commercial programs are easy to use and do not require the user to be an expert in gamma spectrometry. This may lead to problems because even the best programs occasionally give inaccurate results without user intervention. For example, users may not be making all the necessary geometry or attenuation corrections, or checking that the proper nuclide library is being used.

In 1994, synthetic spectra were generated using a computer code developed at the Pacific Northwest Laboratory. Calibration, sample and background spectra were created and converted into a variety of formats which could then be read by most software programs. The synthetic spectra will test peak detection, deconvolution of doublets, and the ability of the program to accurately quantify the nuclides in a complex fission spectrum. These synthetic spectra will be supplied annually to laboratories performing EM analyses. The laboratories will then submit their results to EML for assessment and publication. The results of this program will be incorporated into the EM-263 Integrated Performance Evaluation Program (IPEP) as another tool for evaluation of data quality (see Summary No. 7.3).

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7.8 MIXED ANALYTE PERFORMANCE EVALUATION PROGRAM (MAPEP)

Catherine S. Klusek, Colin G. Sanderson, Isabel M. Fisenne Pamela Greenlaw, Anne Berne, Steven Minick, JoAnn Morris, Pamela Perry, William Rivera, William Rosa, Ada Kong and Yulin L. Tan

DOE's Mixed Analyte Performance Evaluation Program (MAPEP) is administered by DOE's Radiological and Environmental Science Laboratory (RESL), ID. The program will evaluate DOE contractor performance in analyzing typical radionuclide, inorganic and organic analytes found in mixed waste samples at the DOE complex cleanup sites. MAPEP was mandated by a policy directive from the Assistant Secretary for EM in 1994. EML is participating in MAPEP as a first tier laboratory, three in total, which evaluate, test and certify mixed analyte samples for distribution. EML is currently providing radiological measurements and inorganic analyses.

The second MAPEP pilot sample was a soil prepared from a blend of soils from Idaho Nuclear Engineering Laboratory (INEL), ID. The radiological constituents of the soil sample consisted of ^{137}Cs, ^{60}Co, ^{90}Sr, ^{238}Pu, $^{239,240}Pu$, ^{241}Am, ^{244}Cm, and uranium. The hazardous components were the 21 inorganic constituents of the EPA Target Analyte List. The verification round was completed in January 1994, and samples were shipped to 12 participating laboratories in March 1994. The pilot was conducted to test the preparation, distribution and evaluation design of MAPEP (Chem-Nuclear Geotech, 1993). The program will be implemented after evaluation and needed modifications based on the results of the original pilot of a mixed radiological-inorganic water sample and this most recent soil sample.

MAPEP will distribute samples twice a year, in June and December. Initially, the samples will alternate between a water and a soil sample. The first implementation round of MAPEP is scheduled for January 1995. The sample (MAPEP-94-W2) will be a mixed radiological and inorganic water sample. EML completed the verification round analysis in August 1994. The samples were analyzed for five radionuclides (^{137}Cs, ^{57}Co, ^{60}Co, ^{90}Sr and $^{239,240}Pu$), and 11 priority inorganic pollutants.

Preparations are in progress to provide support to MAPEP for the verification of organic analyte concentrations when mixed organic-radioactive samples are added to the program in FY1996. A project plan has been developed to demonstrate proficiency in the testing of volatile organic constituents utilizing standard EPA methods and measurement techniques.

Reference

Chem-Nuclear Geotech, Inc.
"Work Plan for Mixed-Waste Performance Evaluation Pilot Soil Sample IPEP-93-3"
Grand Junction Projects Office Report P-GJPO-855, January 1993, revised November (1993).

7.9 EM-263 ASSESSMENT PROGRAM

Hemant Pandya, Michael Johnson, Vivian Pan and Catherine S. Klusek

EM-263 has developed a comprehensive assessment program for the technical and QA management activities of EM environmental sampling and analysis facilities, external laboratories and field operations to ensure that data collection operations comply with DOE QA requirements and laboratory-specific QA/QC requirements, and that sampling and analysis plans are being effectively implemented. The assess

ment program also evaluates EM sampling and analysis activities to ensure that the approved QA management systems are implemented and effective. The QA management phase of the assessment program was developed during 1994 and was tested in a pilot program. The pilot assessment is described in Section 7.10. After evaluation and incorporation of modifications to the protocol based on the experience with the pilot, EM-263 will implement QA assesments of EM sampling and analysis in 1995.

Team member Support. EML and RESL developed and provided training at EML to potential team members for the assessment program. This is described in detail in Summary No. 7.10. In addition, EML hosted a training program by EPA EMSL-LV for on-site laboratory evaluations of environmental laboratories. This course is used by EPA to train its auditors for monitoring laboratory performance in the Contract Laboratory Program. Additional training was provided as "auditors-in-training" during audits sponsored by other organization, such as Brookhaven National Laboratory (BNL) and the New Jersey Department of Environmental Protection (NJDEP). This participation also provides information regarding how other organizations perform their technical and managerial audits.

A database of qualified assessment team members from EML, RESL and support contractors was developed. The database includes pertinent information such as education, experience, specialty and subspecialty assessment areas. The database was used to identify qualified personnel to participate as technical and managerial assessment team members for the pilot assessment of Site 300 of the Lawrence Livermore National Laboratory (LLNL).

7.10 PILOT ASSESSMENT

Hemant Pandya, Vivian Pan, Richard Larsen, Matthew Monetti and Lambros Kromidas

EML-263 conducted a pilot assessment to readiness test the design of the QA management phase of its Assessment Program. The assessment addressed technical aspects of EM environmental sampling and analysis activities (ground water and soil sampling), and the management QA structures that support Site 300 of Lawrence Livermore National Laboratory, Livermore, CA. The assessment was conducted November 29 through December 2, 1994 with a team composed of seven member from EML and RESL. The assessment team used the guidelines described in the draft "Protocol Document for EML-263 Assessments" USDOE (1994a). The Draft Assessment Report for the Pilot Assessment was issued to EM-263 (USDOE, 1994b).

References

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USDOE EM, Analytical Services Division DOE Assessment Program for EM Environmental Sampling and Analysis Activities: Assessment Report for the Pilot Assessment" Draft Document (1994b)

7.11 ASSESSMENT TEAM DEVELOPMENT - TEAM MEMBER TRAINING COURSE

Michael Johnson, Catherine S. Klusek, Vivian Pan, Hemant Pandya and Jenny May

In October of 1994, a training course was given to potential team members of EM-263's QA Assessment Program. The purpose of the course was to provide orientation towards DOE/EM's concept of quality environmental data collection in support of EM program goals and the EM-263 Assessment Protocol. Participants in the training included technical specialists and management personnel from EML and RESL.

In support of the training, formal course documents (USDOE, in press) were developed at EML during the summer of 1994 to address concepts of the EM-263 Assessment and to complement those concepts introduced in the EM-20 Auditor/Lead Auditor Training Course at DOE Headquarters. The course was designed as a 3 day tutorial covering all aspects of a technical and management assessment. The training material covered areas of EM organization and QA requirements, concepts and philosophy of assessment, the EM-263 assessment program and related protocols, and interview conduct and techniques. The course focused mainly on the training of technical specialists for assessments using established DOE Guidance Documents DOE/0158P and DOE/0159P (USDOE, 1994a,b). Potential QA management team members were trained with the established Performance Objectives and Criteria in Management Assessment Guidance Document, DOE/EM-0161P (USDOE, 1994c).

Five attendees of the course participated in the second pilot of the EM-263 QA Assessment in November of 1994 for Site 300 of the LLNL remediation program. Additional presentations of the training course will be made at EML and RESL in 1995 (USDOE, in press).

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USDOE EM, Analytical Services Division "EM-263 Assessment Training Course" Report USDOE EML-370, in press

7.12 DIRECT SUPPORT TO EM-263

Donald C. Bogen, Catherine S. Klusek, Michael Johnson and Hemant Pandya

The EML staff provides direct support to EM-263 through its membership on the QA Working Group. Through this group, EML provides technical assistance to EM-263 through presentations at meetings, preparation of presentation materials, and frequent visits to EM-263 to work on projects of timely interest to Analytical Services Division (ASD). Additional support is provided through review of documents related to other aspects of EM-263 which are under the Resource Management and Methods and Instrumentation Programs. Activities of the IPEP Working Group are discussed in Summary No. 7.3.

EML continued its participation in EM Environmental Sampling and Analysis Coordinating Committee (ESACC). ESACC is composed of appropriate EM-20, EM-30, EM-40, EM-50 and EM-60 program management and technical staff. The committee provides on-going assistance to EM-263 by identifying significant issues related to sampling and analysis activities requiring coordinated Headquarters resolutions and provides a forum for EM-263 to present its program to the EM program managers. This group meets periodically throughout the year.

Methods and Instrumentation Program. The EML staff acts as reviewers for the DOE Methods Compendium which provides analytical methods that have been developed or modified for use in sampling and analysis of radioactive, mixed waste and other samples unique to the DOE Complex (Goheen, 1994). The document emphasizes methods that are not currently provided by existing manuals, such as EPA SW-846 Methods.

Resource Management Program. The QA Working Group is working closely with the Resource Management Section on areas of performance evaluation and assessment concerns. The National Sample Tracking System databases provide information on resources and sample load that will be utilized in the assessment site selection process.

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7.13 CALIBRATION OF PACKARD TRI-CARB 2250CA LIQUID SCINTILLATION ANALYZER FOR CONVENTIONAL AND CERENKOV COUNTING

Salvatore C. Scarpitta and Isabel M. Fisenne

A commercially available liquid scintillation counter was calibrated for 30 nuclides that emit either alpha, beta, gamma or conversion electrons. Those nuclides which emit beta particles in excess of 263 MeV were counted in either 10 mL of water or a dilute solution of 7-Amino 1,3 Naphthalene diSulfonic Acid (ANSA) to determine the Cerenkov counting efficiency (CCE). Cerenkov counting is an adjunct to liquid scintillation counting that does not require a scintillation cocktail (Kessler, 1966; Haberer, 1966; Takiue, 1993). ANSA is a wavelength shifter which can be added to aqueous solutions to increase the CCE by about 10%. Some nuclides which can be measured by Cerenkov counting are ^{32}P, ^{35}S, ^{36}Cl, ^{40}K, ^{59}Fe, ^{89}Sr, $^{90}Sr/^{90}Y$, $^{106}Rh(Ru)$, ^{137}Cs, ^{210}Bi, and ^{234m}Pa (progeny of ^{238}U via ^{234}Th).

Cerenkov and liquid scintilation measurements were made using carrier free aqueous solutions of ^{3}H, ^{14}C, ^{40}K, ^{45}Ca, ^{54}Mn, ^{55}Fe, ^{59}Fe, ^{99}Tc, ^{95m}Tc, ^{85}Sr, ^{89}Sr, $^{90}Sr/^{90}Y$, ^{133}Ba, ^{137}Cs, $^{106}Ru/^{106}Rh$, ^{207}Bi, $^{210}Bi/^{210}Po$, ^{226}Ra, ^{232}U, natural U, ^{237}Np, ^{241}Am, ^{243}Am, ^{242}Pu and ^{244}Cm. All radionuclides were obtained as standards from the US Department of Commerce National Institute of Standards and Technology (NIST) and Amersham International. Most of the nuclide solutions were calibrated at EML by gas proportional counting (an article will be prepared for submission to a peer reviewed journal).

Standardized solutions, containing the equivalent of about 17 Bq (1000 dpm), were dispensed gravimetrically, in triplicate, into either 20-mL plastic or low borosilicate glass vials. Each vial was counted for 15-30 min with window settings of 0-50 keV for Cerenkov, and 1-2000 keV for liquid scintilation counting so that the 1 sigma counting error was <2%. The liquid scintilation samples were recounted three additional times after the addition of increasing amounts of nitromethane, a chemical quenching agent. The liquid scintilation detection efficiencies at each quench value were determined from the known activities added to each vial that contained 15 mL of chilled Insta-Gel-XF (Packard Instrument Co.) liquid scintillation cocktail. The data were analyzed by regression and utilized to construct quench calibration curves (i.e., counting efficiency versus degree of quenching). The average instrument background, using 20 mL plastic vials containing 3-18 mL of ultrapure water was 0.225 $\pm$ 0.018 counts per second (cps) for a 0-50 keV region of interest . The average background count rate for glass vials (0.346 $\pm$ 0.010 cps) was about 30% higher than that obtained using plastic vials. Based on replicate background measurements, the lower limit of detection (LLD) for a 1 hr count at the 95% confidence level, using water as a solvent, was 0.024 cps and 0.028 cps for plastic and glass vials, respectively.

The optimum CCE was obtained using 10 mL of 25 mM ANSA. Figure 7.4 shows that the CCE increased linearly as beta energies increased from 0.300 to 3.54 MeV, achieving a maximum value of 80% for $^{106}Ru/^{106}Rh$ (B^-_{max} = 3.54 MeV). Using ^{40}K (B^-_{max} = 1.32 MeV), the Cerenkov count rate in ANSA was linear over a 3 orders of magnitude range in beta activity (10 - 10,000 mBq). The LLD for a 1 hr count, using a 25 mM ANSA solution, was 0.039 cps for plastic vials. Based on the LLD in ANSA, the minimum detectable activity for Cerenkov producing radionuclides, assuming a 50% CCE and 50% yield of the nuclide of interest, is about 160 mBq for a 1 hr count.

Figure 7.5 shows the LS counting efficiencies obtained using microliter amounts of tracer added to low ^{40}K borosilicate glass vials containing 15 mL of neat Insta-Gel-XF scintillation cocktail. A comparison of the data showed that the CCE was 20-50% less than the liquid scintilation counting efficiency for beta particles with maximum energies in excess of 1 MeV.

References

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"Measurement of Beta Activities in Aqueous Samples Utilizing Cerenkov Radiation"
Packard Technical Bulletin No. 16, Packard Instrument Co., Downers Grove, IL, January 1966

Kessler, M.
"Cerenkov Counting"
Packard Application Bulletin No. 7, Packard Instrument Company, Downers Grove, IL, December 1986

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"Reliability of Activity Determined by Cerenkov Measurements in a Liquid Scintillation Counter"
J. E. Noakes, F. Schonhofer and H. A. Polack (Editors)
in: Liquid Spectrometry (1992)

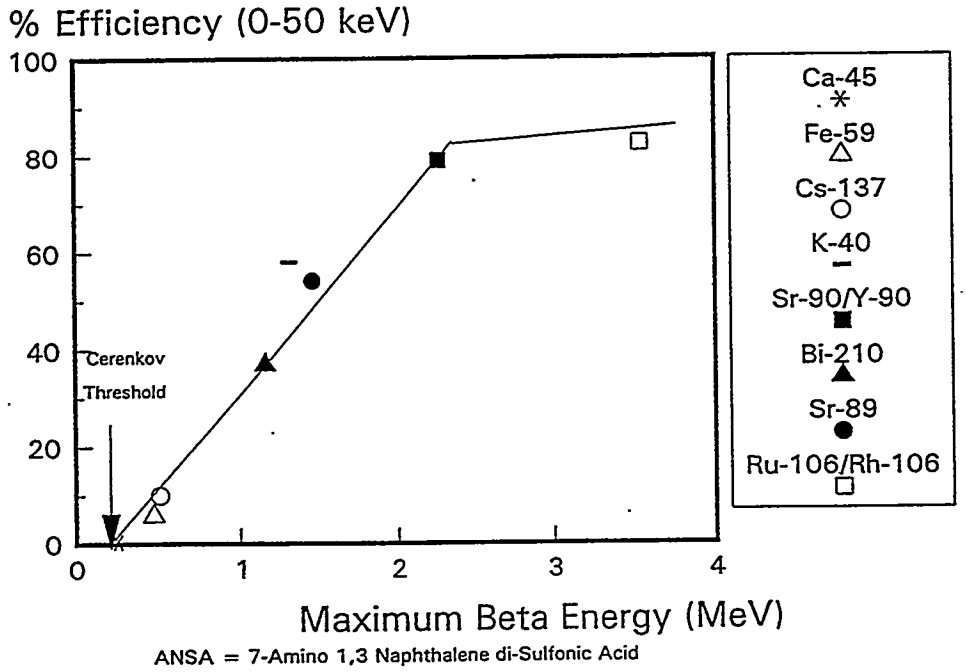

Figure 7.4 Cerenkov Efficiency in 10 mL of 25 mM ANSA.

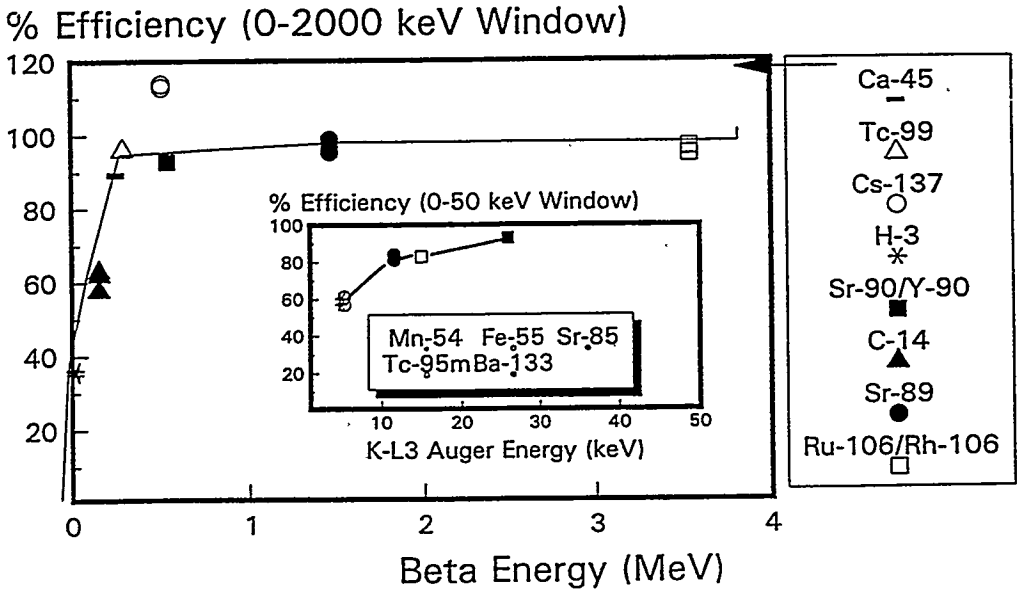

Figure 7.5 Liquid scintilation counting efficiencies in 15 mL Insta-Gel.

7.14 DETERMINATION OF ^{55}FE AND ^{59}FE BY DUAL-DPM MODE LIQUID SCINTILLATION ANALYSIS

Salvatore C. Scarpitta

A radiochemical separation and dual-isotope counting technique for ^{55}Fe has been developed and is being tested for inclusion in the HASL-300 Procedures Manual (in press). ^{55}Fe decays by electron capture to ^{55}Mn (stable) and can only be detected by liquid scintilation counting. The proposed method can separate ^{55}Fe/^{59}Fe in aqueous samples that contain various mixed alpha, beta and gamma emitting radionuclides. The procedure is intended for the determination of ^{55}Fe in QAP samples but is applicable to environmental samples. Beta emitting ^{59}Fe (E_{max} = 0.475 MeV) is added to the samples as the yield determinant prior to $FeOH_3$ scavenging. An identical vial containing ^{59}Fe and carrier is prepared as a reference standard which is later counted to determine the recovered ^{59}Fe.

Following two anion exchange separations that sequentially remove most alpha, beta and gamma interferences, the samples are reduced to a 5 mL volume and converted to $FePO_4$ to minimize color quenching (ASTM 1990). After the addition of 15 mL Insta-Gel cocktail, the samples are counted in a commercially available liquid scintillation counter that is operated in the "Dual-dpm" mode (DeFilippes, 1994). A single count in a calibrated instrument provides the quench corrected activity concentrations of both ^{55}Fe in a sample and recovered ^{59}Fe.

References

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ASTM Committee D19, Philadelphia, PA, March (1990)

DeFilippis, S.
"^{55}Fe and ^{59}Fe: A Qualitative Comparison of Four Methods to Liquid Scintillation Activity Analysis"
Radioactivity Radiochem., 5,14-21 (1994)

"EML Procedures Manual"
US DOE Report HASL-300, 28th Edition, in press

7.15 RAPID DETERMINATION OF ^{90}Sr IN MIXED RADIOACTIVE SAMPLES BY CERENKOV COUNTING

Salvatore Scarpitta

A rapid counting technique was developed in a cooperative study with Tser Min Chang, Institute of Nuclear Research, Taiwan, China, and tested to quantify ^{90}Sr (E_{max} = 0.546 MeV) via its beta emitting progeny, ^{90}Y (E_{max} = 2.27 MeV), in aqueous solutions by Cerenkov counting. This technique is intended for the measurement of aqueous samples that may contain ^{90}Sr with mixed alpha, beta and gamma emitting radionuclides. Cerenkov counting is applicable for beta particles with maximum energies greater than 0.263 MeV. Alpha emitting nuclides will not be detected.

Strontium carrier (30 mg) is added as a gravimetric yield determinant to a known amount, M_s (g) of sample. Yttrium-90 is then separated from the ^{90}Sr by either oxalate precipitation (Procedure Sr-04, HASL-300) or Eichrom Tru-Spec extraction chromatographic resin (Eichrom Industries, Inc., Darrien, IL). An oxalate separation may result in the trapping of interfering radionuclides, whereas the Tru-Spec separation is specific for strontium, allowing most nuclides to elute from the resin bed.

Immediately following separation of ^{90}Y from strontium, a baseline count rate, C_b (counts min^{-1}), is obtained on the Sr fraction, using a commercially available LSC to quantify any Cerenkov contribution by other beta or gamma nuclides that may be present in the sample. The sample is recounted at any three time intervals (h), t_1, t_2, and t_3, over a 48 h period to obtain the corresponding count rates (counts min^{-1}) CT_1, CT_2 and CT_3 as during ingrowth of ^{90}Y above the baseline value. Strontium-90 activity is calculated, from the difference in 90Y count rates above baseline, using the following:

$$A_i 90\ (dpm\ g^{-1}) = \frac{(CT_i - CT_1) - C_b}{M_s \times Eff \times [\, e^{-L\,\Delta t_1} - e^{-L\,\Delta t_i}\,]}$$

where:

i = *interval from separation to the i^{th} count,*
Eff = ^{90}Y Cerenkov counting efficiency, counts min^{-1} dpm^{-1},
L = decay constant for ^{90}Y = 0.01083 h^{-1},
Δt_1 = interval from separation to the 1st count, and
Δt_i = interval from separation to the i^{th} count (1 = 2 or 3).

After the samples are counted, strontium is precipitated as the carbonate, filtered, dried and weighed. The ratio of the recovered weight to that of a reference standard precipitated in the same manner is the yield recovery factor. The average ^{90}Sr value obtained using equation 1 is then corrected by the yield recovery factor.

The Cerenkov counting efficiency for ^{90}Y in water is about 70%, and can be obtained by counting a NIST traceable $^{90}Sr/Y$ solution in secular equilibrium that contains sufficient activity (1000 dpm in 0.1 mL^{-1}) to obtain a <1% counting error.

The Cerenkov counting efficiency for ^{90}Sr is <0.3% for a 0-15 keV counting window. A wavelength shifter, ANSA (7-Amino 1,3 Napthalene di-Sulfonic Acid), can be used to enhance the Cerenkov counting efficiency by about 10% but is not recommended if strontium yield recovery is to be determined gravimetrically.

Samples are typically counted in 20 mL plastic scintillation vials containing 5 - 20 mL of water. For a 20 minute count time, the detection limit is 6 Bq L^{-1} (0.35 dpm mL^{-1}) with a relative standard deviation less than 3%. The method is being tested using a series of QAP water samples containing mixed radionuclides.

References

Chieco, N. A., D. C. Bogen and E. O. Knutson (Editors)
"EML Procedures Manual"
USDOE Report HASL-300, 27th Edition, February (1992)

7.16 EVALUATION OF ASTM COMMITTEE-D19 URANIUM IN DRINKING WATER PROCEDURE USING PERALS

Salvatore C. Scarpitta*

A proposed ASTM procedure (ASTM, 1994) for natural uranium in water was evaluated at EML using high resolution alpha-liquid scintillation spectrometry. The testing protocol developed at EML, consisted of: a) measurements of three USEPA, EMSL-LV, Nevada samples by the proposed ASTM method using PERALS (photon electron rejection alpha liquid scintillation spectrometry, McDowell and McDowell, 1994); b) inclusion of a yield recovery sample spiked with 143 dpm of ^{232}U, and c) inclusion of a mixed alpha standard sample containing 2 dpm mL^{-1} of ^{232}U, ^{243}Am, ^{244}Cm and ^{242}Pu in 100 mL of deionized water. The PERALS spectrometer was initially calibrated with a NIST traceable ^{226}Ra source. The system was checked using 5000-8000 dpm of natural U, ^{232}U, ^{243}Am, ^{244}Cm and ^{242}Pu. The uranium results obtained by PERALS spectrometry were verified by measuring fractions of the unknown water samples, both before and after ASTM radiochemistry, using a Packard Tri-Carb-2250 CA liquid scintillation counter and Insta-Gel XF scintillation cocktail. The liquid scintillation alpha counting efficiencies for both natural uranium and ^{232}U were 2.00 and 1.40 counts min^{-1} dpm^{-1} over a wide range of quenching values. Prior to PERALS counting, we extracted actual test samples with 2.0 mL of a commercially available extractant (URAEX) specific for uranium. We counted 1.0 mL of the extract for 20 min per sample by PERALS, and 0.5 mL of the extract by liquid scintillation 15 mL of Insta-Gel XF for 10 min.

To test the extraction efficiencies of various transuranic alpha emitters into URAEX, we prepared synthetic salt solutions, to which was added ^{232}U, ^{242}Pu, ^{243}Am or ^{244}Cm. We were able to extract 100% of the added ^{232}U tracer into 2 mL of URAEX. We also found that ^{242}Pu and ^{243}Am were extracted into URAEX with extraction efficiencies of 76% and 18%, respectively. ^{244}Cm was not extracted into URAEX.

In general, we found good agreement on the same samples between the liquid scintillation counting post-chemistry counts on the URAEX extracts and the PERALS counts. Yield corrections were made from the internal standards incorporated into our test protocol. The yield recovery, using ^{232}U, was about 72%. However, the liquid scintillation counts obtained on pre- and post-chemistry water samples were not consistent with each other, suggesting trapping of uranium by either the mineral salts present in the Nevada water or Fe complexing. For the liquid scintillation counting post-chemistry samples, the salts were separated from the water, whereas the salts were not separated in samples extracted into URAEX and counted by PERALS.

Based on our evaluation of the ASTM method, we recommended to the Committee that:

1. The actual water samples be purified by either Dowex 4x8 or Tru-Spec columns (Eichrom Industries Inc., Darien, IL) prior to URAEX extraction to remove both the salts and plutonium/americium from the uranium fraction,

2. A ^{232}U yield tracer be incorporated into the ASTM uranium procedure in the event that the extraction efficiency into URAEX is not 100%, and that the equation used to calculate natural uranium be adjusted accordingly, and

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* Project was carried out in conjunction with Tser-Min Chang of the Institute of Nuclear Research, Republic of China.

3. The PEARLS instrument be calibrated with a mixed uranium standard containing 2000 dpm or greater of ^{232}U, ^{234}U and ^{238}U, rather than ^{226}Ra which does not extract into URAEX. Any two peaks in the uranium could be used to calibrate the instrument for energy.

References

McDowell, W. J. and B. L. McDowell, "Liquid Scintillation Alpha Spectrometry" CRC Press, Boca Raton, FL (1994)

ASTM "Standard Test Method for Uranium in Drinking Water and Other Waters by High Resolution Alpha Liquid Scintillation Spectrometry" ASTM Committee D-19 on Water, Draft 5, December (1994)

EM-50 Projects

7.17 FIELD INVESTIGATION AT CHELYABINSK

Karen A. Stevenson

In September 1994, a Russian - American team conducted hydrogeological, geochemical, geophysical and radiometric measurements in the territory of the Mayak Production Association, 13 km southwest of Chelyabinsk-65 (Ozorsk) in the vicinity of the Mishelyak River. The purpose of this demonstration was to examine the frontal area of a groundwater plume moving from Lake Karachai toward the river. Lake Karachai and this region have been contaminated as the result of past accidents and radioactive waste disposal practices (Stevenson, 1994).

This demonstration was conducted under the "Program of Joint Russian - American Field Studies", in the framework of a joint Russian - American agreement between the Russian Ministry of Atomic Energy (MINATOM) and the DOE/EM. Participating U.S. laboratories included the Environmental Measurements Laboratory (EML), Lawrence Berkeley Laboratory (LBL) and Pacific Northwest Laboratory (PNL). Activities conducted at the field site included the collection of groundwater from selected depths using both American and Russian well samplers, a surface electrical resistivity and induced polarization survey using a dipole-dipole resistivity array, radiometric measurements along the Mishelyak River, and a visual geological survey of nearby rock exposures.

The EML team members (Karen A. Stevenson and Wayne Lowder, consultant) were tasked with providing the American team with both real-time and passive dosimetry and conducting *in situ* gamma-ray spectrometry measurements along the Mishelyak River. These field radiometric measurements were made using a small pressurized ion chamber for environmental radiation (SPICER), that was designed and built at EML, and a 5 cm x 5 cm Na(Tl) portable "SCOUT" spectrometer, supplied by Quantrad Sensor Incorporated, USA. The average free-air absorbed dose at the field site was approximately 25 $\mu R\ h^{-1}$. Figure 7.6 depicts an *in situ* gamma-ray spectra that was obtained along a transverse at the field site. As shown, the predominant gamma-ray emitting radionuclides are ^{137}Cs and ^{40}K.

At the conclusion of this joint field demonstration, a preliminary report titled "On Joint Russian - American Hydrogeological and Geophysical Research which took place on 9-19th, September, 1994 in the Area of the Production Association MAYAK Site was prepared and presented to the Vice-Minister of MINATOM, N. N. Egorov, in

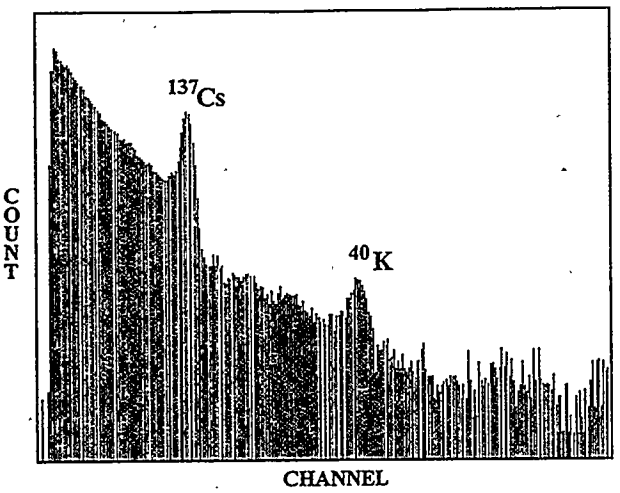

Figure 7.6 Gamma-ray spectra obtained at a field test site adjacent to MAYAK. Peaks depicted in figure are of the ^{137}Cs 662 keV and ^{40}K 1461 keV emission lines.

Moscow. A detailed report of the American team's findings will be published in 1995 as a DOE Headquarters document.

References

Stevenson, K. A.
"Some Aspects of Radioactive Contamination Within the Former USSR"
in: Managing Radioactive and Mixed Wastes, Proceedings of the Twenty Seventh Mid-Year Topical Meeting of the Health Physics Society, Albany, New York, pp. 337-380, July (1994)

7.18 REGIONAL AND GLOBAL TRENDS IN BACKGROUND AIR CONCENTRATIONS OF URANIUM AND PLUTONIUM FOR DOE WASTE REMEDIATION MONITORING

Karen A. Stevenson and Vivian Pan

Within the U.S. regulators and public interest groups are pressuring Federal agencies to accelerate the cleanup of contaminated facilities. Often, a complicating factor in characterizing areas contaminated with very low levels of radioactivity is differentiating the fraction attributable to the source in question from natural background. In an effort to discern regional differences in background uranium and plutonium surface air concentrations, EML has requested historical databases from DOE's Argonne National Laboratory (ANL) - East's Environmental Monitoring Program, Battelle Pacific Northwest Laboratory (PNL), and EPA's Environmental Radiation Ambient Monitoring System (ERAMS).

An assessment of the uranium (Stevenson and Pan, in press) and plutonium (Pan and Stevenson, in press) surface air data from the late 1970s to the present has been made. The historical total activity levels for both radionuclides were shown to fall within the range of concentration levels that have been observed in Europe. The current plutonium concentration levels are 1000 times lower than had previously been recorded in the 1960s (Perkins and Thomas, 1980). This drastic decline in plutonium concentration levels is a direct result of the global ban on atmospheric weapons testing. Over this same time period, the concentration levels of uranium in the northern and mid-section of the country have shown a

decrease of approximately 40%. This regional trend of decreasing surface air uranium concentration levels may be attributed to regulatory compliance in reducing emissions from fossil fuel burning facilities. A mean average $^{234}U/^{238}U$ activity ratio within the U.S. was determined to be 1.1 (0.3 1s). For these ratio values, there was no evidence of any historical trends, seasonal changes, regional effects or coastal variations.

$^{234}U/^{238}U$ and $^{235}U/^{238}U$ data reflect a mixing of natural background uranium with a maximum of 1% high-enriched uranium.

The post-fallout concentration of $^{239,240}Pu$ have remained at detectable concentrations close to conventional minimum detection limits, whereas ^{238}Pu is now below detection limits. Upon inspection of this post-fallout time period, it is

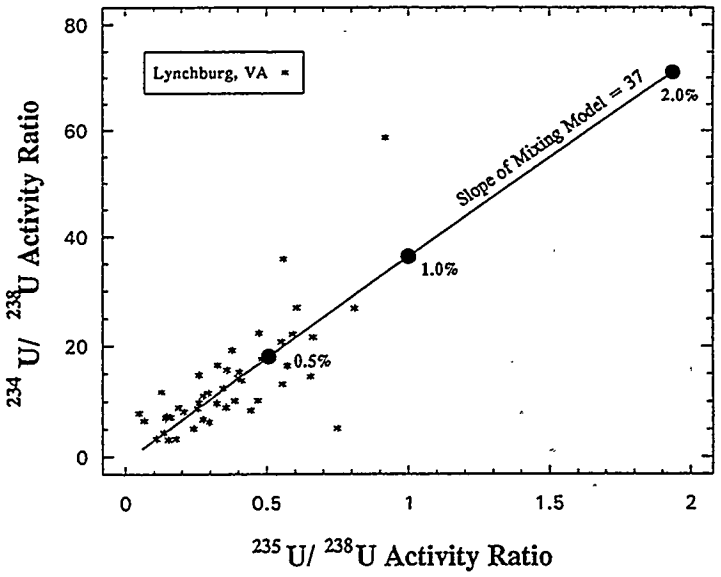

Figure 7.7 A mixing line curve of the $^{234}U/^{238}U$ and $^{235}U/^{238}U$ activity ratio results for a mixing model of natural uranium and high-enriched source superimposed with results of the EPA ERAMS Lynchburg database (*). Contours of the percent high-enriched source are marked by dots (●)on the mixing line.

A simple mixing model was developed to assess what fraction of the uranium observed at the ERAMS Lynchburg air sampling station could be attributed to a local source. Figure 7.7 shows a plot of this mixing model for background and a high-enriched uranium source with contours of the percent of high-enriched sources (dots) on the mixing line, and the Lynchburg data superimposed over the model. A regression of the Lynchburg

apparent that the surface air concentrations of $^{239,240}Pu$ vary over a 10-fold range. Further resolution can be obtained through an analysis of the historical data in a temporal context. As shown in Figure 7.8 for the ANL site 12N data and the PNL data, one can describe these temporal variations with the first derivative of concentration with respect to time $\delta C/\delta t$, where C is the monthly average concentration and t is in units

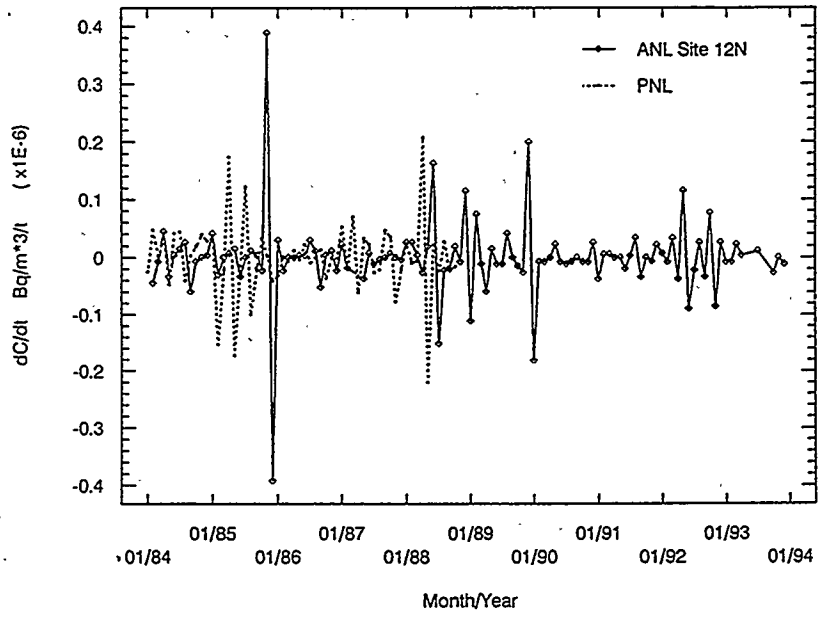

Figure 7.8 History of the monthly changes of $^{239,240}Pu$ in surface air from 1984-1993 as shown by $\delta C/\delta t$ vs. time, where C is in units of μBq m^{-3} and t is 1 month. Data are for the ANL site 12N and PNL.

of 1 month. Results are useful for environmental baseline interpretation because this analysis allows one to determine with 95% confidence whether a future observed concentration is within acceptable baseline fluctuation limits.

References

Pan, V. and K. A. Stevenson
"Temporal Variations Analysis of Plutonium Baseline Concentration in Surface Air From Selected Sites in the Continental U.S."
EML Report, in press

Perkins, R. W. and C. W. Thomas
"Worldwide Fallout"
in: *Transuranic Elements in the Environment*, W. C. Hanson (Editor),
Report USDOE/TIC-22800, pp. 53-82 (1980)

Stevenson, K. A. and V. Pan
"An Assessment of Uranium in Surface Air Within the Continental U.S."
J. of Environ. Radioactivity, in press

7.19 TECHNICAL PROGRAM MANAGER (TPM) ACTIVITIES FOR THE OFFICE OF TECHNOLOGY DEVELOPMENT (EM-50)

Karen A. Stevenson

The DOE's Office of Technology Development (EM-50) has been given the mission to direct an aggressive national campaign to accelerate the development of innovative technologies to remediate contaminated DOE sites. The responsibility of the Technical Program Manager (TPM) under this initiative is to be the site single point of contact for headquarters oversight.

During this past year, Karen Stevenson, the TPM, for EML again served as a technical focus group committee member for the selection of Technical Task Plans (TTP) for the Characterization, Monitoring and Sensor Technology Crosscutting Program (CMST-CP) under the EM Focus Area initiative. Dr. Stevenson also acted in the capacity of technical expert for the Office of Demonstration Testing and Evaluation (EM-54) in discussions with Auburn Steel Company concerning the characterization of ^{137}Cs contaminated "bag house" dust.

8

STAFF ACTIVITIES

- Seminar Program
- Academic Affiliations and Activities
- Editorial Activities
- Committee and Outside Coordination Activities (National)
- Professional Society Activities
- Standards Activities
- Committee and Outside Coordination Activities (International)
- Meetings / Programs Organized
- Visiting Scientists
- Publications
- Publications in Press
- Presentations

SEMINAR PROGRAM

The seminar program at EML serves two purposes: to keep the scientific staff informed of progress in major programs within the Laboratory and to keep us up to date on programs in other institutions through visiting scientists. The speakers and topics listed below were presented during 1994.

Stuart Nagourney, New Jersey Department of Environmental Protection
"Analytical Chemistry and Environmental Regulatory Policy: Friend or Foe?"
March 29

Gladys Klemic, EML
"Results of 10th International Intercomparison of Environmental Dosimeters"
April 5

Burton G. Bennett, UNSCEAR
"Radiation Exposures World-wide"
April 19

Adam R. Hutter, EML
"Soil Gas Thoron/Radon Ratios"
May 3

Earl O. Knutson, EML
"New Radon Chamber at EML"
May 10

Lambros Kromidas, EML
"Electron Microscopy Investigations at EML"
June 21

Volker Genrich, Genitron Instruments, Germany
"Radon and Climatic Multiparameter Analysis: An Effective Tool for Optimization and Verification of Radon-Reduction Techniques"
July 29

Al Cavallo, EML
"The Industrial-Scale Development of Wind Energy"
August 9

Richard J. Larsen, EML
"EML's Global Radioactivity Sampling Network"
September 13

Laszlo Koblinger, KFKI Atomic Energy Research Institute, Hungary
"Overview of Activities of Health Physics Department, KFKI" and "Migration of Cesium Originating from Chernobyl Fallout in Various Soil Types, 1987-1993"
November 4

Awards

Isabel M. Fisenne

Achievement Award from the American Society for Testing and Materials (ASTM) Committee C26.05 on the Nuclear Fuel Cycle, presented in Santa Fe, NM

Achievement Award for distinguished contributions in the field of radioanalytical chemistry, presented at the 40th Conference on Bioassay, Environmental and Analytical Chemistry, Cincinnati, OH

Academic Affiliations and Activities

Herbert W. Feely

Member, Thesis Advisory Committee, City University of New York

Isabel M. Fisenne

Adjunct Associate Professor, Nelson Institute of Environmental Medicine, New York University

Andreas C. George

Member, Thesis Advisory Committee, Rutger's University, New Jersey

Carl V. Gogolak

Lecturer, Harvard University Short Course Program, School of Public Health
Adjunct Lecturer, Mathematics Department, Polytechnic University

Paul Goldhagen

Adjunct Assistant Professor, Center for Radiological Research, Columbia University

S. Frederic Guggenheim

Stevens Institute of Technology
Member, Executive Committee and Decade Director, Alumni Association
Member, Old Guard Committee
Founding Member and Member, Executive Committee, Alumni Environmental Professionals

Merrill Heit

Member, Thesis Advisory Committee, St. Johns University

Adam Hutter

Adjunct Lecturer, City University of New York

Earl O. Knutson
Adjunct Associate Professor, Institute of Environmental Medicine, New York University

Lambros Kromidas
St. John's University
Adjunct Assistant Professor, College of Pharmacy and Allied Health
Member, Thesis Adivsory Committee, Toxicology Department

Robert Leifer
Member, Thesis Advisory Committee, Brooklyn College
Associate Member, Graduate Faculty of Rutgers University

Kevin M. Miller
Lecturer, Harvard University Short Course Program, School of Public Health

Vincent C. Negro
Adjunct Assistant Professor, Queensboro Community College

Marcel Reginatto
Adjunct Instructor, The Cooper Union for the Advancement of Science and Art

Keng-Wu Tu
Member, Thesis Advisory Committee, New York University

Editorial Activities

The senior staff peer reviewed 45 articles in response from editors of many different technical journals, proposals submitted to the Department of Energy, ER and OHER, as well as other agencies supporting research.

Committee and Outside Coordination Activities (National)

Harold L. Beck
Chair, National Council on Radiation Protection and Measurements (NCRP), Scientific Committee 93, Radiation Measurements
Member, NCRP, Scientific Committee 84, Bulk Contamination
Chair, NCRP, Scientific Committee 84-1, Surface Soil Contamination

Donald C. Bogen
- Member, Federal Interagency Task Group on "National Handbook of Recommended Methods for Water Data Acquisition"
- Member, Technical Subcommittee #1, Siting, Chemistry and Quality Assurance, National Atmospheric Deposition Program

Alfred A. Crescenzi
- Member, ER Occupational Safety and Health Quality Improvement Team

Herbert W. Feely
- Member, Technical Committee, National Atmospheric Deposition Program (NADP)

Andreas C. George
- Member, DOE Radiological Control Committee's Subcommittee on Occupational Exposure to Radon
- Member, American Waterworks Association's Advisory Committee for the Critical Assessment of Radon Progeny Exposure While Showering with Radon Bearing Water

Carl V. Gogolak
- Chair, Environmental and Public Radiation Protection Committee, Council on Ionizing Radiation Measurement and Standards

Ferenc Hajnal
- Member, Advisory Committee on Shielding Requirements for Deep Space Mission, NASA, Langley Research Center
- Member, Radiation Health Review Panel to NASA, American Institute of Biological Sciences

Merrill Heit
- Key EML Representative to OHER Environmental Sciences Division (ER-74)
- DOE Representative, Interagency Arctic Research Policy Coordinating Committee (IARPC)
- DOE Advisor, to DoD-Office of Naval Research Arctic Research Progrm
- Member, Interagency SERDP EcoRisk Methodology Working Group

Michael J. Johnson
- Member, QA Working Group, DOE/EM-263, Office of Compliance and Site Coordination
- Member, Mixed Analyte Performance Evaluation Program Working Group, DOE/EM-263, Office of Compliance and Site Coordination
- Member, Integrated Performance Evaluation Programs Working Group, DOE/EM-263, Office of Compliance and Site Coordination

Catherine S. Klusek
Member, QA Working Group, DOE/EM-263, Office of Compliance and Site Coordination
Member, Mixed Analyte Performance Evaluation Program Working Group, DOE/EM-263, Office of Compliance and Site Coordination
Member, Integrated Performance Evaluation Programs Working Group, DOE/EM-263, Office of Compliance and Site Coordination

Earl O. Knutson
Member, Air Sampling Instruments Committee of the American Conference of Government Industrial Hygienists

Robert Leifer
Member, Interagency Steering Group on Airborne Geoscience
Member, Aerosol Team, Atmosphere Radiation Measurement Program, DOE

Camille Marinetti
Secretary, EEO/Federal Women's Program Subcommittee (New York Federal ExecutiveBoard)

Kevin M. Miller
Organizer, IEEE Short Course, "*In Situ* Gamma-Ray Spectrometry for Site Characterization," 1994 Nuclear Science Symposium
Member, Review Panel, DOE-OHER Measurement Science Program

Hemant S. Pandya
Member, QA Working Group, ASD Programs DOE/EM-263, Office of Compliance and Site Coordination

Professional Society Activities

Donald C. Bogen
Member, Health Physics Society, Inter-Society Committee on Methods of Air Sampling and Analysis

Carl V. Gogolak
Health Physics Society:
President-Elect, Radiation Section
Chair, National Program Committee
Member, Board of Directors, Radon Section

S. Frederic Guggenheim
American Society of Mechanical Engineers (ASME):
Member,Education Committee, Design Engineering Division
Liaison, Education Committee, Design Engineering Division to Board, Engineering Education
Member, Region 2 Operating Board: Professional Development
Member, Board, Engineering Education, Ad Hoc Committee, Membership Task Force

Robert Leifer
Member, Advisory Committee, New York Academy of Sciences, Meteorological Section

Kevin M. Miller
Session Chair, Health Physics Society 1994 Annual Meeting

Vivian Pan
Member, American Geophysical Union, Committee on Public Affairs
Member, American Institute of Physics, Committee on Public Policy

Standards Activities

Isabel M. Fisenne
Member, Committee on Nuclear Fuel Cycle, Task Group on Environmental Methods, American
Society for Testing and Materials, ASTM C-26.05

Andreas C. George
Member, Radionuclides Task Group, Subcommittee on Indoor Air Quality, American Society for Testing and Materials, ASTM D22.05
Member, Working Group, Performance Specifications for the Measurement of Radon in Indoor Air, American National Standards Institute (ANSI) ANSI-N13.34

Michael Johnson
Member, Subcommittee on Sampling Methods, ASTM D34.01
Chair, Subcommittee on Screening Methods, ASTM D34.02.06
Member, Subcommittee on Mixed Waste, ASTM D34.09
Member, Subcommittee on Environmental Regulations, ASTM E50.04
Member, Subcommittee on Sampling Methods, ASTM D18.01

Gladys A. Klemic
Member, Working Group, Standard for Environmental Dosimeters, ANSI-N13.37
Member, Working Group, Standard for Environmental Dosimetry Performance Criteria for Testing, ANSI-N13.29

Hemant S. Pandya
Member, Subcommittee on Environmental Regulation Performance Standards, ASTM E50.04

Colin G. Sanderson
Chair, Subcommittee on Radiochemical Methods of Analyses for Water and Water Deposits, ASTM D19.04
Member, Subcommittee on Radioactivity Standards, ANSI-N1042.2

Committee and Outside Coordination Activities (International)

Nestor Azziz
Technical Program Consultant, Seventh International Symposium on Thermoluminescent Dosimetry, Mexico, September 1994

Harold L. Beck
United States Delegate, International Electrotechnical Commission (IEC) Technical Committee TC-45, Scientific Committee SC-45B, Member Working Groups B5 and B10
Member, Working Group 7.1, Environmental Transport, of Joint US - USSR Coordinating Committee on Civilian Nuclear Reactor Safety

Isabel M. Fisenne
Member, Organizing Committee, International Intercalibration Intercomparison Program (IIIP) for Radon, Thoron and Daughters Measuring Equipment, Nuclear Energy Agency (OECD) and Commission of the European Communities (CEC)

Andreas C. George
Member, Organizing Committee, International Intercalibration Intercomparison Program (IIIP) for Radon, Thoron and Daughters Measuring Equipment, Nuclear Energy Agency (OECD) and Commission of the European Communities (CEC)
Member, International Atomic Energy Agency Coordinated Research Program for International Radon Metrology Program

Merrill Heit
U. S. Delegate, International Arctic Monitoring Assessment Program (AMAP), U. S. State Department

Gladys A. Klemic
Corresponding Member, European Radiation Dosimetry Group (EURADOS) Working Group, Environmental Radiation Monitoring

Catherine S. Klusek
- Member, Research Coordination Group, International Atomic Energy Agency, Research Program on Rapid Instrumental and Separation Methods for Monitoring Radionuclides in Food and Environmental Samples
- Member, Research Coordination Group, International Atomic Energy Agency, Research Program on Development and Selection of Analytical Techniques for Measuring Accidentally Released Radionuclides in the Environment

Earl O. Knutson
- Member, Fuchs Memorial Awards Committee, International Aerosol Research Assembly
- Member, Organizing Committee, International Intercalibration Intercomparison Program (IIIP) for Radon, Thoron and Daughters Measuring Equipment, Nuclear Energy Agency (OECD) and Commission of the European Communities (CEC)

Kevin M. Miller
- Member, Report Committee on *In Situ* Gamma Spectrometry, International Commission on Radiation Units and Measurements

Colin G. Sanderson
- Member, International Committee for Radionuclide Metrology, Low-Level Group

Karen Stevenson
- Member, Program of Joint Russian-American Field Studies (DOE/EM)

Meetings Organized

Donald C. Bogen
- 12th Annual DOE Analytical Managers Meeting, October 4-6

Carl V. Gogolak
- 39th Annual Meeting of the Health Physics Society, San Francisco, CA, July 26-30

Visiting Scientists

As always, during the past year EML received a great many official visitors from all corners of the world, generally for periods of just a day or two. The following individuals worked/trained at EML for longer periods:

Christian Lange
- RISO National Laboratory, Denmark

Amaal Amad Tawik
Research Fellow, Nuclear Research Centre, Radiation Protection Department, Atomic Energy Authority, Cairo, Egypt

Tser-Min Chang
Institute of Nuclear Research Taiwan

Tae-Soon Park
Korea Research Institute of Standards and Science, Republic of Korea

EML PUBLICATIONS
1/01/94 - 12/31/94

Albert, B. : See No.(s)28

1. Azziz, N., G. A. Klemic and R. Fried
"Measurement of Thermal Neutrons in Mixed (n, gamma) Fields Using Aluminum Oxide TLDs"
Nuclear Science Journal, Vol. 31, pp. 59-63, February, 1994

Azziz, N. : See also No.(s)15, 16

Beasley, T. M. : See No.(s)24

Beck, H. L. : See No.(s)13, 20

2. Chen, C. J., P. S. Weng, T. C. Chu and E. O. Knutson
"Discrimination of Airborne Artificial Radioactivity from Radon Progeny"
Health Physics, Vol. 66, pp. 557-564, May, 1994

3. Cheng, Y. S., C. C. Yu and K. W. Tu
"Intercomparison of Activity Size Distributions of Thoron Progeny by Alpha-and Gamma-Counting Methods"
Health Phys., Vol. 66, pp. 72-79, January, 1994

Chieco, N. A. : See No.(s)13

Decker, K. M. : See No.(s)20

4. Fisenne, I. M.
"Uranium"
H. G. Seiler, A. Sigel, H. Sigel (Editors)
in: Handbook on Metals in Clinical and Analytical Chemistry, Chapter 56, Marcel Dekker, Inc., New York, pp. 639-649, April, 1994

5. Fisenne, I. M.
"Lead-210 in Animal and Human Bone: A New Analytical Method"
Environment International, Vol. 20, pp. 627-632, August, 1994

6. Fisenne, I. M., A. C. George, P. M. Perry and H. W. Keller
"The April 1993 and November 1993 Radon Intercomparisons at EML"
USDOE Report EML-562, July, 1994

7. George, A. C. and E. O. Knutson
"Particle Size of Unattached Radon Progeny in Filtered Room Air"
Radiation Protection Dosimetry, Vol. 56, pp. 119-121, December, 1994

George, A. C. : See also No.(s)6, 11, 12, 29, 35

Gogolak, C. V. : See No.(s)12

Goldhagen, P. : See No.(s)15, 16

Hajnal, F. : See No.(s)15, 16

EML PUBLICATIONS
1/01/94 - 12/31/94

Heit, M. : See No.(s)9

8. Huffert, A. M., R. A. Meck and K. M. Miller
"Background as a Residual Radioactivity Criterion for Decommissioning - Appendix A to the Generic Environmental Impact Statement in Support of Rulemaking on Radiological Criteria for Decommission of NRC-Licensed Nuclear Facilities"
Draft Report NUREG-1501, U. S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research, August, 1994

9. Kada, J., M. Heit and K. M. Miller
"Chronologies of Anthropogenic Trace Element Input to Four Utah Lakes Reconstructed Using Sediment Cores"
Water, Air and Soil Pollution, Vol. 75, pp. 353-369, June, 1994

Keller, H. W. : See No.(s)6

Klemic, G. A. : See No.(s)1, 16, 26

Klusek, C. S. : See No.(s)30, 31

Knuth, R. H. : See No.(s)22

Knutson, E. A. : See No.(s)29

10. Knutson, E. O.
"Random and Systematic Errors in the Graded Screen Technique for Measuring the Diffusion Coefficient of Radon Decay Products"
J. Aerosol Sci., Suppl. 1, Vol. 25, pp. 71-72, January, 1994

11. Knutson, E. O. and A. C. George
"Measurements of Lead-214 Loss by Recoil from Decay of Polonium-218 Collected on a Wire Screen"
J. Aerosol Sci. -Proceedings Edition, Vol. 25, pp. 71-72, December, 1994

12. Knutson, E. O., A. C. George, P. Shebell and C. V. Gogolak
"EML Thoron Gas Measurements"
Radiation Protection Dosimetry (Proceedings Issue), Vol. 56, pp. 263-266, December, 1994

Knutson, E. O. : See also No.(s)2, 7, 35

13. Krey, P. W. and H. L. Beck
"EML Annual Report -Calendar Year 1993"
N. A. Chieco (Editor)
USDOE Report EML-560, April, 1994

EML PUBLICATIONS
1/01/94 - 12/31/94

14. Krey, P. W., M. S. Feiner, C. G. Sanderson, J. McInroy, K. G. Inn and J. M. Hutchinson
"A Bone Ash Standard for Strontium-90, Lead-210, Polonium-210, Uranium and the Actinides"
Radioanalytical and Nuclear Chemistry, Vol. 177, pp. 5-18, March, 1994

15. Kugel, H. W., G. Ascione, S. Elwood, J. Gilbert, L. P. Ku, J. Levine, K. Rule, N. Azziz, P. Goldhagen, F. Hajnal and P. Shebell
"Measurements of TFTR D-T Radiation Shielding Efficiency"
in: Fusion Engineering and Design, Proceedings of the Third International Symposium on Fusion Nuclear Technology, Los Angeles, Ca, September, 1994

16. Kugel, H. W., J. Gilbert, D. Hwang, M. Lewis, J. Levine, L. P. Ku, K. Rule, F. Hajnal, N. Azziz, P. Goldhagen, G. A. Klemic and P. Shebell
"TFTR Radiation Contour and Shielding Efficiency Measurements During D-D Operations"
in: Fusion Technology, Proceedings of the Eleventh Topical Meeting on the Technology of Fusion Energy, American Nuclear Society, September, 1994

17. Larsen, R. J. and C. G. Sanderson
"Radioactivity in the Surface Air at Cape Grim"
A. L. Dick, J. L. Gras (Editors)
Baseline Atmospheric Program, Australia, 1991, Department of the Environment, Sport and Territories, Bureau of Meteorology, May, 1994

18. Larsen, R. J. and C. G. Sanderson
"Radioactivity in the Surface Air at BRW, MLO, SMO, and SPO During 1993"
J. T. Peterson, R. M. Rosson (Editors)
Climate Monitoring and Diagnostics Laboratory No. 22, Summary Report 1993, NOAA, U. S. Department of Commerce, December, 1994

19. Larsen, R. J., H. N. Lee, M. Monetti and C. G. Sanderson
"EML'S Global Sampling Program"
Proceedings of the Conference on Atmospheric Chemistry, American Meteorological Society, January 23-28, 1994, pp. 180-186, January, 1994

20. Larsen, R. J., C. G. Sanderson, H. N. Lee, K. M. Decker and H. L. Beck
"Fission Products Detected in Alaska Following the Tomsk-7 Accident"
Journal of Environmental Radioactivity, Vol. 23, pp. 205-209, April, 1994

Larsen, R. J. : See also No.(s)32

Latner, N. : See No.(s)32

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1/01/94 - 12/31/94

21. Lee, H. N. and R. Leifer
"Backward Air Mass Trajectory Analysis for the First CART Site at Lamont, Oklahoma"
Proceedings of the Third Atmospheric Radiation Measurement (ARM) Science Team Meeting, Conf-9303112, UC-402, pp. 357-358, March, 1994

Lee, H. N. : See also No.(s)19, 20, 22

22. Leifer, R., R. H. Knuth and H. N. Lee
"Surface Aerosol Measurements at Lamont, Oklahoma"
Proceedings of the Third Atmospheric Radiation Measurement (ARM) Science Team Meeting, CONF-9303112, UC-402, pp. 349-351, March, 1994

Leifer, R. : See also No.(s)21

23. Maiello, M. L.
"Long-Term TLD Environmental Radiation Measurements at Sites in the New York City Metropolitan Area"
USDOE Report EML-563, November, 1994

24. Mann, L. J. and T. M. Beasley
"Iodine-129 in the Snake River Plain Aquifer at the Idaho National Engineering Laboratory, Idaho, 1990-1991"
U.S. Geological Survey Water-Resources Investigations Report 94-4053 (DOE/ID-22115), April, 1994

25. Miller, K. M.
"Large-Area Proportional Counter for In Situ Transuranic Measurements"
Transactions of the American Nuclear Society, Vol. 70, pp. 47-48, July, 1994

26. Miller, K. M., P. Shebell and G. A. Klemic
"In Situ Gamma-Ray Spectrometry for the Measurement of Uranium in Surface Soils"
Health Physics, Vol. 67, pp. 140-150, August, 1994

Miller, K. M. : See also No.(s)8, 9

27. Monetti, M.
"Strontium-90 Deposition at SMO and MLO Through 1990"
J. T. Peterson, R. M. Rosson (Editors)
Climate Monitoring and Diagnostics Laboratory No. 22, Summary Report 1993, U. S. Department of Commerce, December, 1994

Monetti, M. : See also No.(s)19

Perry, P. M. : See No.(s)6

EML PUBLICATIONS
1/01/94 - 12/31/94

28. Polito, M. and B. Albert
"A Low Power, Constant Air Flow System"
USDOE Report EML-558, January, 1994

29. Reineking, A., E. A. Knutson, A. C. George, S. B. Solomon, J. Kesten, G. Butterweck and J. Porstendorfer
"Size Distribution of Unattached and Aerosol-Attached Short-Lived Radon Decay Products: Some Results of Intercomparison Measurements"
Radiation Protection Dosimetry, Vol. 56, pp. 113-118, December, 1994

30. Sanderson, C. G. and C. S. Klusek
"Semi-Annual Report of the Department of Energy, Office of Environmental Restoration and Waste Management, Quality Assessment Program"
USDOE Report EML-559, January, 1994

31. Sanderson, C. G. and C. S. Klusek
"Semi-Annual Report of the Department of Energy, Office of Environmental Restoration and Waste Management, Quality Assessment Program"
USDOE Report EML-561, July, 1994

32. Sanderson, C. G., N. Latner and R. J. Larsen
"Environmental Gamma-Ray Spectroscopy at Remote Sites with Satellite Data Transmission"
Nuclear Instrum. Methods in Physics Research A, Vol. 339, pp. 271-277, March, 1994

Sanderson, C. G. : See also No.(s)14, 17, 18, 19, 20

Shebell, P. : See No.(s)12, 15, 16, 26

33. Stevenson, K. A.
"Some Aspects of Radioactive Contamination Within the Former USSR"
Managing Radioactive and Mixed Wastes, Proceedings of the Twenty-Seventh Mid-Year Topical Meeting of the Health Physics Society, pp. 377-380, July, 1994

34. Tan, Y. L. and K. W. Tu
"Ultra-High-Volume Sampling of Airborne Particle Associated Polycyclic Aromatic Hydrocarbons"
Proceedings of the 14th International Symposium on PAH, Vol. 5, pp. 185-192, August, 1994

35. Tu, K. W., E. O. Knutson and A. C. George
"Thoron versus Radon: Comparison of Measured Progeny Aerosol Size Distributions"
Aerosol Science and Technology, Vol. 20, pp. 266-274, March, 1994

Tu, K. W. : See also No.(s)3, 34

EML PUBLICATIONS
1/01/94 - 12/31/94

36. Van Steveninck, W.
"A Pressurized Ion Chamber Monitoring System for Environmental Radiation Measurements Utilizing a Wide Range, Temperature Compensated Electrometer"
IEEE Proceedings of Nuclear Science Symposium, Vol. 41, pp. 1080-1085, August, 1994

PUBLICATIONS IN PRESS

Albert, B.
"Light Experimental Aircraft Aerosol Measurement Package"
USDOE Report

Collé, R., M. P. Unterweger, J. M. R. Hutchinson, S. Whittlestone, G. Polian, B. Ardouin, J. G. Kay, J. P. Friend, B. W. Blomquist, W. Nadler, T. T. Dang, R. J. Larsen, and A. R. Hutter
"Part II: Results for the Participating Laboratories"
Journal of Geophysical Research

EML Procedures Manual
28th Edition, Vol. I
USDOE Report HASL-300, in press*

Fisenne, I. M.
"Quality Assurance Procedures and Requirements for the Operation of an Environmental Radiological Measurements Performance Evaluation Program"
NIST: Workshop on Measurement Quality Assurance for Ionizing Radiation

Fisenne, I. M. and P. M. Perry
"Depleted Uranium Exposures Compared to Natural Uranium Environmental Exposures"
Proceedings of 40th Annual Bioassay, Analytical and Environmental Radiochemistry Meeting, Cincinnati, OH

George, A. C.
"State-of-the-Art of Instruments for Measuring Radon/Thoron and their Progeny in Dwellings.--A Review"
Health Physics

George, A. C., K. W. Tu, and E. O. Knutson
"Intercomparison of Active and Passive Instruments for Radon and Radon Progeny in North America"
USDOE Report EML-566

George, A. C., K. W. Tu, and E. O. Knutson
"Third Indoor Radon and Radon Progeny Measurements Intercomparison of Instruments and Methods"
Proceedings of the International Workshop on Indoor Air - An Integrated Approach

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*Projected publication date late 1995 or early 1996.

Gogolak, C. V.
"Non-Parametric Statistical Methodology for the Design and Analysis of Final Status Decommissioning Surveys"
USNRC Report NUREG-1505

Gubala, C. P., D. H. Landers, M. Monetti, M. Heit, T. Wade, B. Lasorsa, and S. Allen-Gil
"The Rates of Accumulation and Chronologies of Atmospherically Derived Pollutants in Arctic Alaska, USA"
The Sci. Total Environ.

Hutter, A. R.
"A Method for Determining Soil Gas ^{220}Rn (Thoron) Concentrations"
Health Physics

Johnson, M.
"Selection and Use of Equipment for the Sampling of Liquids"
USDOE Report

Klemic, G.
"USDOE's International Intercomparisons of Environmental Dosimeters"
Proceedings 6th International Intercomparisons of Environmental Dosimeters

Krey, P. W. and C. S. Klusek
"Soil Sampling in Complex Terrain"
J. Radioanalytical and Nuclear Chemistry

Kromidas, L. and R. Leifer
"An Innovative Application of a Commercially Available Double-sided Adhesive for the Collection of Aerosols by Impaction"
Atmospheric Environment

Landers, D. H., J. Ford, C. Gubala, M. Monetti, B. K. Lasorsa, and J. Martinson
"Mercury in Vegetation and Lake Sediments from the U. S. Arctic"
Water, Soil, Air Pollut.

Larsen, R. J. and C. G. Sanderson
"Radionuclide Results in Air Filter Samples, 1992"
Commonwealth Scientific and Industrial Research Organization, Cape Grim Baseline Air Pollution Station, Baseline 92 Report

Larsen, R. J., M. Monetti, H. N. Lee, and C. G. Sanderson
"EML's Global Sampling Program"
Proceedings of the Conference on Atmospheric Chemistry, American Meteorological Society

Miller, K. M.
"Proposed Methodologies for Measuring Low Levels of Residual Radioactivity for Decommissioning"
USNRC Report NUREG-1506

Olsen, C., T. Beasley, R. J. Larsen, M. Monetti, and M. Heit
"Measurement of Radiation in the Alaskan Atmospheric, Terrestrial and Marine Environments"
Proceedings of the Workshop on Arctic Contamination, Anchorage, AK

Pan, V.
"Analysis of EML QAP Data from 1982-1992: Determination of Operational Criteria and Control Limits for Performance Evaluation Purposes"
USDOE Report

Sanderson, C. G., V. Pan, and P. Greenlaw
"Semi-annual Report of the Department of Energy, Office of Environmental Management, Quality Assessment Program"
USDOE Report

Scarpitta, S.
"A Theoretical Model for Radon-222 Adsorption on Activated Charcoal Canisters in Humid Air Based on Polanyi Potential Theory"
Health Physics

Tan, Y. L., A. Kong, M. A. Monetti, B. Lau, C. Gubala, and D. Landers
"Polycyclic Aromatic Hydrocarbons and Polychlorinated Dibenzo-p-dioxins and Dibenzofurans in Sediment from a Sub-Arctic Lake in Alaska"
13th International Symposium on Chlorinated Dioxins and Related Compounds

Wilson, J. W., J. E. Nealy, F. A. Cucinotta, J. L. Shinn, F. Hajnal, M. Reginatto, and P. Goldhagen
"Radiation Safety Aspects of Commercial High-Speed Flight Transportation"
NASA Technical Paper 3524

EML PRESENTATIONS* From: 01/01/94
To: 12/31/94

Beck, H. L.
"Contemporary Data Bases for ORERP Dose Reconstruction"
NATO/SCOPE-RADTEST Advanced Research Workshop on Atmospheric Nuclear Tests, Vienna, Austria, January 11, 1994

Beck, H. L.
"The ORERP County Data Base"
39th Annual Health Physics Meeting of the Health Physics Society, San Francisco, CA, June 28, 1994

Berne, A.
"Tru-Spec and Teva-Spec Used to Separate Thorium and Neodymium from Americium after Microprecipitation"
40th Annual Bioassay, Analytical and Environmental Radiochemistry Meeting, Cincinnati, OH, November 14, 1994

Bogen, D. C.
"Status of DOE's QA Programs"
NIST Measurement Standard Materials for Studies of Radionuclides in Oceans Workshop, Gaithersburg, MD, June 1, 1994

Fisenne, I. M. and P. M. Perry
"Depleted Uranium Exposures Compared to Natural Uranium Environmental Exposures"
40th Annual Bioassay, Analytical and Enviromental Radiochemistry Meeting, Cincinnati, OH, November 16, 1994

George, A. C.
"State-of-the-Art Instruments for Measuring Radon/Thoron and Their Progeny in Dwellings"
39th Annual Health Physics Meeting of the Health Physics Society, San Francisco, CA, June 28, 1994

George, A. C.
"Overview of EML's Radon Progeny Particle Size Measurement Techniques Covering the Range from 0.5 nm - 5000 nm"
Queensland University of Technology Workshop on Particle Size Characterization, Australia, November 26, 1994

George, A. C.
"Third Intercomparison of Instruments and Methods for Measuring Radon and Radon Progeny in Indoor Air"
Indoor Air - An Integrated Approach Conference, Gold Coast of Australia, November 29, 1994

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* ERRATUM

The presentation "Organics in Alaskan Lake Sediments" reported last year should have had as the presenters: Tan, Y. L., A. Kong, and M. Heit.

George, A. C.
"Radon and Radon Progeny Intercomparison Results from the April and October 1994 EML Exercises"
IAEA Meeting on the Coordinated Research Program - Radon in the Human Environment, Vienna, Austria, December 14, 1994

Gogolak, C. V.
"Non-Parametric Statistics for Decommissioning Surveys"
Meeting of Multi-Agency Radiation Site Investigation Manual (MARSIN) Committee, Nuclear Regulatory Commission, Rockville, MD, November 15, 1994

Gogolak, C. V.
"Research Priorities for Environmental Radiation Protection"
Third Annual Meeting of the Council for Ionizing Radiation Measurements and Standards (CCRMS), NIST, Gaithersburg, MD, November 16, 1994

Heit, M.
"An Overview of New EML Research for the OHER Atmospheric Chemistry Program"
Annual Meeting of the Atmospheric Chemistry Program, Nashville, Tennessee, January 19, 1994

Hutter, A. R.
"Rn-222 at Bermuda and Mauna Loa: Local and Distant Sources"
American Nuclear Society's Third International Conference on Methods and Applications of Radioanalytical Chemistry, Kona, Hawaii, April 13, 1994

Hutter, A.
"Profiles and Models of Soil Gas Radon-220 and Radon-222 at Two Sites in New Jersey"
1994 Annual Meeting of the Geological Society of America, Seattle, WA, October 24, 1994

Hutter, A. and C. V. Gogolak
"Reproducibility of Rn-222 Flux Measurements Using the Accumulation Method"
1994 Fall Meeting of the American Geophysical Union, San Francisco, CA, December 6, 1994

Johnson, M.
"Audit Standards for Field Sampling and Field Measurement"
Tenth Annual Waste Testing and Quality Assurance Symposium, Arlington, VA, July 13, 1994

Klemic, G.
"Results of the 10th International Intercomparison of Environmental Dosimeters"
Riso National Laboratory, Roskilde, Denmark, June 17, 1994

Klusek, C. S.
"QA Audits"
Environmental Sampling and Analysis Coordinating Committee (ESACC) Meeting, Gaithersburg, MD, July 6, 1994

Klusek, C. S.
"EML QAP"
Performance Evaluation Programs Working Group - Laboratory Representatives Joint Meeting, Alexandria, VA, July 13, 1994

Knutson, E. O., K. W. Tu, A. C. George and C. V. Gogolak
"Some Characteristics of the New Radon/Aerosol Chamber in the USDOE Environmental Measurements Laboratory"
Fourth International Aerosol Conference, Los Angeles, CA, September 1, 1994

Krey, P. W.
"Environmental Monitoring"
DOE Laboratory Advisory Group for Effluent Research (LAGER), U. S. Department of Energy, Forrestal Building, Washington, D.C., January 24, 1994

Krey, P. W.
"The Direction of Federal Agencies"
Environmental Issues and the Environmental Scientist, Columbia University, Department of Geological Sciences, New York, NY, February 23, 1994

Landers, D. H., J. Ford, C. Gubala, S. Allen-Gil, B. K. Lasorsa, T. Vlasova, M. Monetti and E. A. Crecelius
"Mercury in Terrestrial and Freshwater Arctic Ecosystems"
International Conference on Mercury as a Global Pollutant, Whistler, B.C., Canada, July 11,1994

Larsen, R. J., H. N. Lee, M. Monetti and C. G. Sanderson
"EML's Global Sampling Programs"
Conference on Atmospheric Chemistry, Nashville, Tennessee, January 25, 1994

Larsen, R. J.
"SASP/RAMP"
Office of Naval Research High Latitude Dynamics, Arctic Environmental Monitoring Strategy, Arlington, Virginia, April 4, 1994

Larsen, R. J.
The Global Environmental Sampling Network of the Environmental Measurements Laboratory
New Directions in Environmental Radioactivity Monitoring, XXXVIIIth IAEA General Conference Special Scientific Programme, Vienna, Austria, September 20, 1994

Larsen, R. J.
"EML Radon Research and NPACS Surface Measurements"
ACP Annual Science Meeting, Seattle, WA, October 19, 1994

Larsen, R. J.
"SASP/RAMP"
Office of Naval Research High Latitude Dynamics, Arctic Environmental Monitoring Strategy, Arlington, VA, April 4, 1994

Larsen, R. J.
"EML Radon Research and NPACS Surface Measurements"
ACP Annual Science Meeting, Seattle, WA, October 19, 1994

Lee, H. N.
"Transport and Diffusion Model Using the Semi-Lagrangian and Spectral Method"
Fourth International Conference on Atmospheric Sciences and Application to Air Quality, Seoul, Korea, June 2, 1994

Leifer, R.
"EML's Radionuclide Database and Aerosol Measurements"
ACP Annual Science Meeting, Seattle, WA, October 19, 1994

Miller, K. M.
"A Large Area Proportional Counter for In-Situ Transuranic Measurements"
American Nuclear Society Annual Meeting, New Orleans, LA, June 22, 1994

Miller, K. M.
"Chernobyl Follow Up: IAEA Environmental Assessment and Preliminary Reports of Observed Health Effects"
Advanced Workshop on Nuclear Emergency Planning, Harvard School of Public Health, Boston, MA, July 27, 1994

Miller, K. M. and P. Shebell
"In Situ Gamma-Ray Spectrometry for Site Characterization"
IEEE Nuclear Science Symposium Short Course, Norfolk, VA, November 2, 1994

Miller, K. M.
"NRC Initiatives"
12th Annual Doe Analytical Manager's Meeting, Montvale, NJ, October 5, 1994

Pan, V.
QAP: New Report/Evaluation
12th Annual DOE Analytical Manager's Meeting, Montvale, NJ, October 4, 1994

Sanderson, C. G., N. Latner and R. J. Larsen
"Remote Atmospheric Gamma-Ray Spectroscopy with HPGe Detectors and Satellite Data Transmission"
36th Rocky Mountain Conference on Analytical Chemistry, Denver, CO, August 1, 1994

Scarpitta, S. C.
"Radon Lung Dose Contribution Due to Showering"
39th Annual Health Physics Meeting of the Health Physics Society, San Francisco, CA, June 29, 1994

Scarpitta, S. C.
"Measuring Technetium-99 by Liquid Scintillation Counting Using a New Extraction Chromatographic Material"
Canberra User's Group Meeting, Charlotte, NC, June 6, 1994

Scarpitta, S. and I. M. Fisenne
"Cerenkov Counting as a Complement to Liquid Scintillation Counting"
40th Annual Bioassay, Analytical and Environmental Radiochemistry Meeting, Cincinnati, OH, November 14, 1994

Shebell, P.
"Application of Pressurized Ionization Chambers and Germanium Spectrometers to Environmental and Neutron Radiation Studies at EML"
Riso National Laboratory, Roskilde, Denmark, June 17, 1994

Stevenson, K. A.
"DOE Analytical Laboratory Integrated Assessment Program"
Technical Information Exchange Meeting with Industry on Environmental Restoration Needs in the Northwest United States, Seattle, Washington, January 26, 1994

Stevenson, K. A.
"Some Aspects of Radioactive Contamination Within the Former USSR"
Twenty-seventh Midyear Topical Meeting of the Health Physics Society, Albany, New York, February 16, 1994

Stevenson, K. A.
"EML's Radionuclide Measurement Capabilities"
Lawrence Berkeley Laboratory Chelyabinsk Workshop, California, May 17, 1994

Stevenson, K. A.
"U.S. Department of Energy - EM Program"
SPECTRUM '94, Atlanta, GA, August 15, 1994

Stevenson, K. A.
"The U.S./Russian Joint Field Studies at Mayak, September 9-19, 1994: Real-Time Dosimetry Measurements and Other Observations"
Ames Laboratory, Iowa, October 27, 1994

Tu, K. W. and E. O. Knutson
"Measurement of Radon Progeny Size Distribution With a MOUDI and a Graded Screen Array"
Fourth International Aerosol Conference, Los Angeles, CA, September 1, 1994

9. ABBREVIATIONS AND ACRONYMS

ACP	Atmospheric Chemistry Program
ALARA	as low as reasonably achievable
AMAP	Arctic Monitoring Assessment Programs
AMS	accelerator mass spectrometry
ANATEX	Across North America Tracer Experiment
ANL	Argonne National Laboratory
ANSI	American National Standards Institute
APG	Aberdeen Proving Grounds
APRF	U. S. Army Pulse Radiation Facility
ARM	Atmospheric Radiation Measurement
ASCOT	Atmospheric Studies in Complex Terrain
ASTM	American Society for Testing and Materials
BCMS	beaded carbon molecular sieve
BNL	Brookhaven National Laboratory
CART	cloud and radiation test bed
CCD	charged coupled device
CCE	Cerenkov counting efficiency
CDIAC	Carbon Dioxide Information Analysis Center
CEC	Commission of European Communities
CMDL	Climate Monitoring and Diagnostics Laboratory
D-D	deuterium-deuterium
DOD	Department of Defense
DOE	Department of Energy
DOELAP	DOE's Laboratory Accreditation Program
D-T	deuterium-tritium
DREO	Defense Research Establishment, Ottawa
DQO	data quality objective
EH	Environment, Safety and Health
EM	Office of Management
EPA	Environmental Protection Agency
EURADOS	European Radiation Dosimetry Group
ERAMS	Environmental Radiation Ambient Monitoring System
ESACC	Environmental Sampling and Analyses Coordinating Committee
FSA	full spectrum analysis
GAW	Global Atmospheric Watch
GMD	geometric mean diameter
GSA	graded screen array
GSD	geometric standard deviation
GW	gross alpha/beta water
HASP	High Altitude Test Chamber
HP	Hewlett-Packard
HPGe	high purity germanium
HSCT	high-speed civil transport
HZE	high charge and energy
IAEA	International Atomic Energy Agency
IARPC	Interagency Arctic Policy Coordinating Committee
ICRU	International Commission on Radiation Units and Measurements
IIIP	International Intercalibration and Intercomparison Program

INEL	Idaho National Engineering Laboratory
IPEP	Integrated Performance Evaluation Program
IRMP	International Radon Metrology Program
LAHET	Los Alamos High Energy Transport Code
LAN	local area network
LBL	Lawrence Berkeley Laboratory
LLD	lower limit of detection
LSC	liquid scintillation counter
MAPEP	Mixed Analyte Program
MARSIM	Multi-agency Radiation Site Investigation Manual
MCA	multichannel analyzer
MINATOM	Russian Ministry of Atomic Energy
MOHAVE	Measurement Of Haze and Visual Effects
MOUDI	micro-orifice uniform deposit impactor
NASA	National Aeronautics and Space Administration
NCI	National Cancer Institute
NIH	National Institute of Health
NIST	National Institute of Standards and Technology
NOAA	National Oceanic and Atmospheric Administration
NRC	Nuclear Regulatory Commission
NSF	National Science Foundation
NTS	Nevada Test Site
NYU	New York University
OHER	Office of Health and Environmental Research
ONR	Office of Naval Research
PAEC	potential alpha concentration
PAH	polycyclic aromatic hydrocarbons
PC	personal computer
PE	performance evaluation
PERALS	photon electron rejection alpha scintillation
PIC	pressurized ionization chamber
PNL	Pacific Northwest Laboratory
PPPL	Princeton Plasma Physics Laboratory
PTB	Physikalisch-Technische Bundesanstalt
QA	quality assurance
QAP	Quality Assurance Program
QA/SAC	Quality Assurance/Science Activity Center for the America Americas
QC	quality control
RAMP	Remote Atmospheric Measurements Program
RAMS	remote atmospheric measurements systems
RARAF	Radiological Research Acceleration Facility
RESL	Radiological Environmental Science Laboratory
RH	relative humidity
RWMC	Radioactive Waste Management Complex
RMC	Royal Military College
RTG	radionuclide thermal generator
SASP	Surface Air Sampling Program
SEM	scanning electron microscopy
SFR	sample flow rate
SMPS	scanning mobility particle sizer
SPICER	small pressurized ion chamber for environmental radiation

TEDE	total effective dose equivalent
TFTR	Tokamak Fusion Test Reactor
TIMS	thermal ionization mass spectrometry
TLD	thermoluminescent dosimetry
TPM	Technical Program Manager
TTP	Technical Task Plan
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
UOM	University of Miami
WMO	World Meteorological Organization