EML-580

ENVIRONMENTAL MEASUREMENTS LABORATORY

1995 annual report

July 1996

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DISCLAIMER

1995 annual report

EML

Environmental Measurements Laboratory

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EML - 580

August 16, 1996

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In order to serve our colleagues and stakeholders in an effective and cost-efficient way, we have made the EML Annual Report for Calendar Year 1995 (EML-580 dated July 1996) accessible in downloadable form from our INTERNET site on the World Wide Web at

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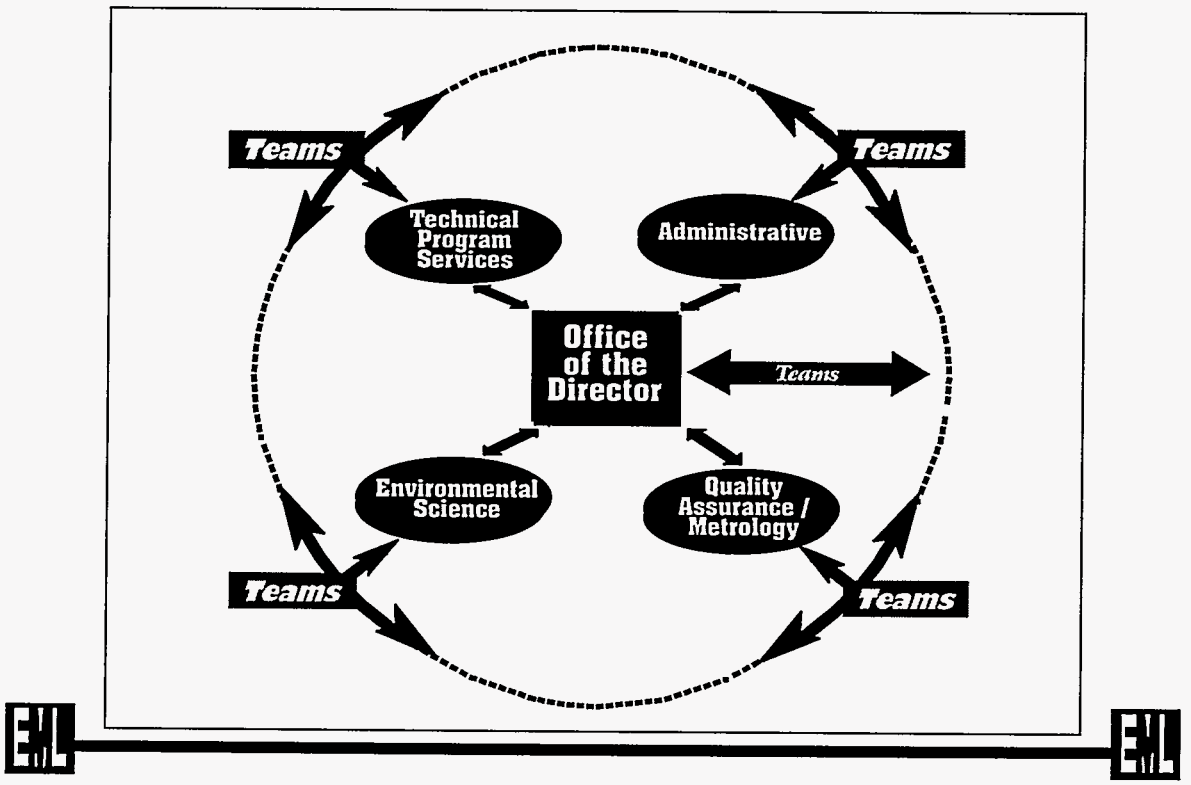
Copies will be available sometime in September from the National Technical Information Service, 5285 Port Royal Road, Springfield, Virginia 22161.

We hope our report will be of interest to you. We welcome your inquiries.

Sincerely,

Philip W. Krey, Acting Director
Environmental Measurements Laboratory

The EML Organization Chart



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1 ENVIRONMENTAL RADIATION AND RADIOACTIVITY

1.1 OVERVIEW

Kevin M. Miller

The Environmental Radiation and Radioactivity program area encompasses a variety of projects principally aimed at collecting and interpreting fundamental data on natural and anthropogenic radionuclides in the environment and the related ionizing radiation field produced. In some cases, the projects lead to associated studies of non-nuclear pollutants. The data collected in EML's programs can be used to model environmental pathways and to estimate the dose and the health impact to humans on a global, regional, or local basis. The hallmark of many of these projects over the years has been a rapid response capability to address important environmental issues that arise within the DOE complex, the United States, or around the globe.

A significant component of this program area includes the operation and maintenance of EML's Global Network, a group of over 115 sampling sites dispersed throughout the world for measuring radioactivity in the air and in precipitation. The Network can assess any new introduction of radioactivity into the environment, including episodic releases associated with clandestine or accidental nuclear weapon detonations as well as with accidents associated with nuclear reactors, the transportation sector, and the launch and reentry of space vehicles containing radioactive materials. Included in the Network are Remote Atmospheric Measurements Systems which feature on-site sample analysis and satellite communication for near real-time assessment of events.

Information from the EML Global Network constitutes a significant portion of the world's surface air database. The Network's records of radionuclide concentrations serve as benchmark data with which to test global circulation models developed for climate investigations. Our experience in sampling and analysis, garnered over four decades of Network operations, and the continued commitment to quality assurance has led to EML being designated as a calibration center for the World Meteorological Organization's Global Atmospheric Watch Program.

In addition to projects relating to atmosphere inventories and deposition to land, EML participates in studies to characterize radionuclide distributions in the Arctic Ocean. This work is part of the Office of Naval Research program to determine if radioactive waste management practices of the Former Soviet Union have potentially compromised fisheries or led to radioactivity levels of concern in the Arctic ecosystem.

Special studies are undertaken at facilities within the DOE complex to evaluate the distribution and cycling of radioactive contaminants. Basic information on the dynamics of the systems under study is required to assess the environmental impacts of remediation strategies and their efficacy in environmental restoration programs. In addition, EML's past work in lake sediment coring as part of localized and regional studies involving radionuclides has lead to spin-off studies that focus on toxic and potentially carcinogenic organic compounds. This work is being performed in conjunction with the U.S. Environmental Protection Agency (EPA) as part of their Dioxin Exposure Initiative and Arctic Contaminant Research Program.

The final element of this program area relates to the assessment of environmental radiation fields produced by both natural background and anthropogenic radionuclides. An ongoing study of external radiation exposure from terrestrial and cosmic sources provides fundamental information for assessing the contributions from nuclear technology as well as for setting release criteria for residual radioactivity at sites undergoing environmental restoration. Quality assessment projects that are international in scope are performed in the area of environmental radiation dosimeters, in particular, thermoluminescence dosimeters. Our program also includes standards work for these devices as they represent a principal means for documenting environmental exposures at DOE and other nuclear facilities.

1.2 GLOBAL RADIONUCLIDE DEPOSITION STUDIES

Matthew A. Monetti, Karin M. Decker, Sylvia Hulse, and William Rivera

EML has been investigating the atmospheric deposition of radioactivity for 40 years, and this scientific pursuit continues to provide information addressing issues of concern within the scientific community. In 1958, the laboratory instituted a global network of sampling sites to determine the global transport and fate of radionuclides released as a result of atmospheric testing of nuclear weapons. A single radiologically important fission product, ^{90}Sr, was measured in the samples collected during the first 32 years of the program. The database generated proved valuable for determining factors controlling the global distribution of radioactive fallout, as well as providing source-term information for studies using anthropogenic radionuclides as biogeochemical tracers of various processes. Since the last atmospheric weapons test in 1980, there has been only one reported large-scale release of anthropogenic radioactivity into the atmosphere, namely the Chernobyl Accident. The deposition of ^{90}Sr was insignificant and primarily undetectable from the early 1980s until 1990 except for 1986, the year of the Chernobyl Accident (Monetti and Larsen, 1991; Monetti, in press). As a result of the lack of any definitive atmospheric source of ^{90}Sr and studies conducted during the past two years, the radionuclide analysis of the deposition samples was changed to gamma spectrometry. The radionuclides of particular interest include naturally-produced isotopes, ^{7}Be and ^{210}Pb, and fission products, ^{137}Cs, ^{95}Zr and ^{144}Ce. Gamma spectrometric measurements of fission products enable the program to continue to be used to identify accidental or intentional atmospheric releases of radioactivity. The ^{7}Be and ^{210}Pb data is of special interest to researchers developing global circulation models since it may be used to verify the models. The data will also be available to calculate the source term and inventories of ^{210}Pb and ^{7}Be for colleagues interested in using these radionuclides as biogeochemical tracers. Since the gamma analysis is non-destructive, the samples are archived or used for additional studies. The data generated in these studies can be directly coupled to those found in the Surface Air Sampling Program (SASP) (see Summary No. 1.3). The integration of the results from these two programs leads to the investigation of the removal processes of these radionuclides from the atmosphere.

At the end of 1995, deposition samples were being collected at the 79 locations shown in Figure 1.1. The sampling station in Columbia, Missouri, was removed from the global network as a result of staffing changes at that location. 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. All of the monthly collections, except for those collected at eight specific locations, are composited into quarterly samples at EML. The samples collected at the other eight sites are analyzed as monthly samples in order to obtain more detailed results of the radionuclide deposition, particularly the short-lived ones. The samples are analyzed by gamma spectrometry and then used for other studies or archived for potential future use. During 1995, these eight sites included Yap Island; Cold Bay, Alaska; Munich, Germany; Columbia, Missouri; Keflavik, Iceland; Bangkok, Thailand; Harare, Zimbabwe; and Guayaquil, Ecuador. These sites were chosen to represent various climatological conditions and a wide global distribution. Special studies of radionuclide deposition were also conducted in 1995 in response to the detonations of underground nuclear weapons by China and France (see Summary No. 1.5).

About 200 samples were processed and analyzed by gamma spectrometry during the past year. Over a quarter of these samples was collected during the Chinese and French testing. These samples were returned to EML by express shipping services to provide us with the opportunity to determine if any short-lived radionuclides were present. The remainder of the analyzed samples was collected as part of the normal studies during 1995. Fission products were below the detection limit in all but four samples. The activity in these four samples was low and temporally and spatially isolated, so they are not likely to represent an unannounced release of anthropogenic radionuclides. The only fission product detected was long-lived ^{137}Cs. Its presence may have been due to resuspension. The natural radionuclides, ^{7}Be and ^{210}Pb, were detected much more frequently, but not in all of the samples. ^{7}Be has a short half-life (53.3 days), so it will only be found in samples analyzed soon after collection. Detectable activities of ^{7}Be were found in 67 samples. Many of the samples analyzed within a few months of collection did not have detectable ^{7}Be activities. ^{210}Pb, however, has a much longer half-life (22.3 years), and this isotope should remain in all the samples that were analyzed. The detector background and matrix blank of ^{210}Pb is a more significant concern for ^{210}Pb measurements. These factors cause the limit of detection for ^{210}Pb and the error associated with ^{210}Pb analysis to be higher. ^{210}Pb was detected in 110 of the samples. Radionuclide deposition based on samples analyzed to date was found to go up to values of 850 and 65 Bq/m^2 per quarter for ^{7}Be and ^{210}Pb, respectively.

The current database of gamma-emitting radionuclide deposition is rather limited, so it is not easily possible to discern any temporal or global trends in the deposition. It is expected that continued studies will reveal these trends. In 1996, attempts will be made to better coordinate these studies with the SASP in order to address some factors involved in the transport and removal of these radionuclides from the atmosphere.

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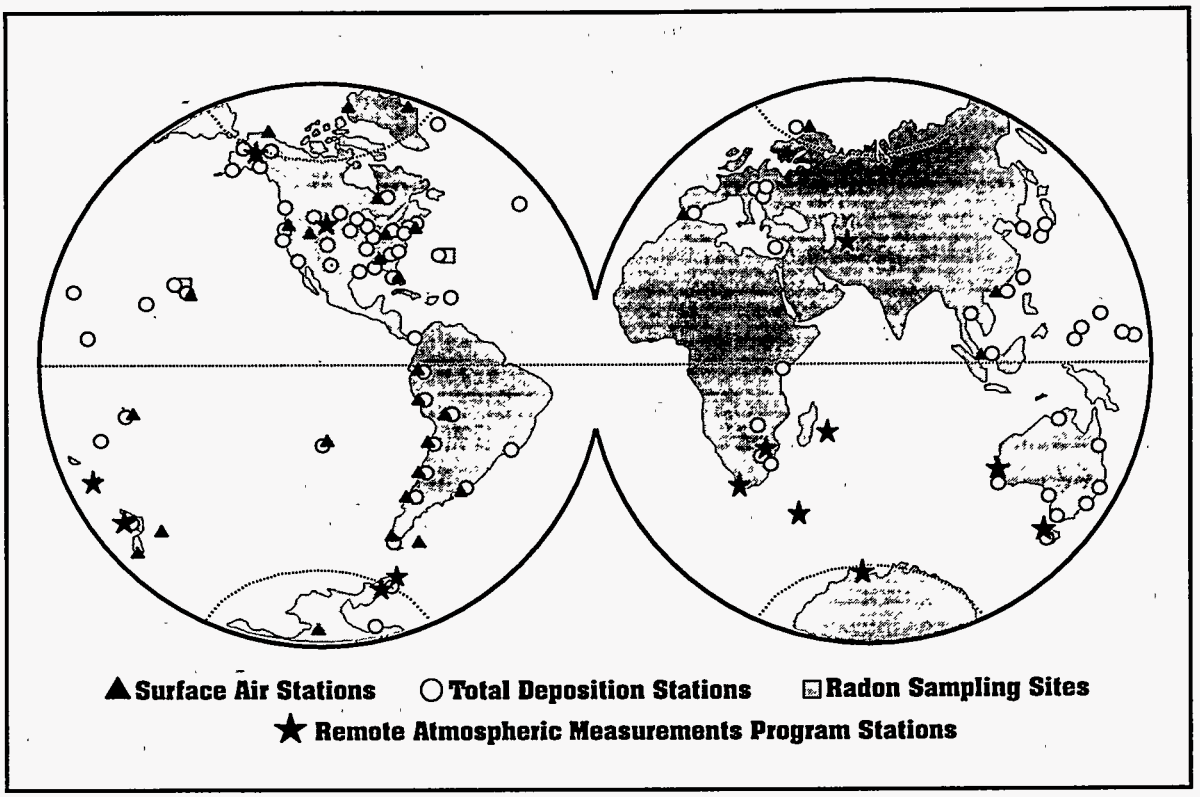

Figure 1.1 EML's Global Sampling Network.

1.3 THE SURFACE AIR SAMPLING PROGRAM (SASP)

John Kada, Colin G. Sanderson, Richard Godwin and William Rivera

Through the Surface Air Sampling Program (SASP), EML operates a network of high volume aerosol samplers at 41 sites around the world. In its early years, the objective of the SASP program was to trace the atmospheric dispersion of radionuclides produced by surface testing of nuclear weapons. SASP also has provided the capability to monitor anthropogenic releases of anthropogenic radionuclides to the atmosphere from accidents, as demonstrated after the 1986 accident at the Chernobyl power reactor and the 1993 accident at the Tomsk-7 facility. In 1995, public concern over a series of subsurface nuclear weapons tests conducted by France at its test sites at Mururoa and Fangataufa Atolls in the Pacific Ocean led to enhanced sampling at several SASP sites in and around the southern Pacific Ocean (see Summary No. 1.5).

The SASP network also provides a platform for the long-term study of the global tropospheric concentration of a number of substances besides artificial radionuclides, and this has become an increasingly important aspect of the SASP program in recent years. SASP regularly measures and reports surface air concentrations of the naturally-produced radionuclides ^{7}Be and ^{210}Pb at all its sites. This data is now widely used in the global climate modeling community to evaluate aerosol transport and scavenging components of global climate models. The recent report of the SASP program (Larsen et al., 1995) adds to this global database of ^{210}Pb and ^{7}Be and includes concentration data for four SASP sites added to the network in 1993: Gibraltar, Singapore, Hong Kong, and Tromso.

The SASP sampling network also serves as a platform for the study of the atmospheric cycles of stable chemical species of interest to scientists involved in climate change and global biogeochemical research. The University of Miami (UOM) analyzes air filter samples from SASP sites in remote regions for a suite of stable chemical species as part of its research on global scale chemistry of the troposphere. Particular emphasis is placed on nitrogen and sulfur species since aspects of the atmospheric chemistry of these species are still not fully understood. Temporal and spatial variations in sulfur and nitrogen concentration in remote areas where anthropogenic influences are minimal provide an opportunity to focus on the natural production, transformation and removal processes affecting these species. ^{210}Pb and ^{7}Be measurements made by SASP at these sites play an important role in the interpretation of the nitrogen and sulfur data by providing clues which help to unravel the effects of transport and aerosol scavenging processes. Because of the important role atmospheric sulfur species are thought to have on the radiative balance of the atmosphere via the "direct" and "indirect" aerosol forcing effects of sulfate aerosol, understanding the atmospheric cycle of these species is also relevant to global climate studies.

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USDOE Report EML-572, November (1995)

1.4 REMOTE ATMOSPHERIC MEASUREMENTS PROGRAM (RAMP)

Colin G. Sanderson, Steven Minick, Karin M. Decker, Norman Latner, Norman Chiu, Vincent C. Negro, Scott Wurms, Camille Marinetti and John Kada

RAMP is an extension and modification of the Surface Air Sampling Program (SASP) (see Summary No. 1.3) and was initiated in 1987. In addition to having the goals of SASP, RAMP, depending upon the communication system being used, provides real-time and 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 analyzes 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 through-put geostationary satellites (METEOSAT), transferred to ground stations, and automatically recovered via a telephone link by EML's computer. The spectra are then automatically reconstructed and analyzed at EML. 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.

In April 1995, a sodium iodide RAMS was installed near Pinedale, WY. This system, similar to the one installed in Alaska in 1994, utilizes land telephone lines for two-way communications between the remote site and EML. Personnel located at EML in New York can now perform the same analysis functions with the RAMS that site personnel perform via these telephone lines.

A new, completely automatic RAMS (AUTORAMP) has completed its laboratory testing at EML. Unattended operation of EML's RAMS is now possible for one month if daily samples are collected, or up to three months if samples are collected on a three-day cycle. With this new system, the gamma-ray detector is a mechanically-cooled high-purity germanium diode coupled to a portable multichannel analyzer (MCA). A portable computer is used to control the MCA, a "pick & place" robot system, and data flow. Sealed lead acid gel-cell batteries and trickle chargers are used for

continuous d.c. current operation. The transmitted spectra and on site peak search analysis are reconstructed and reviewed at EML.

On July 5, 1995, the AUTORAMP (see Figure 1.2) was installed at McClellan AFB, Sacramento, CA, for field testing and evaluation. Although the mechanical functions of the AUTORAMP were operational on July 13, the electrically cooled germanium detector failed. In mid August, another detector was installed and backup sensor switches were added to the system. On August 10, 1995, AUTORAMP became completely operational and began transmitting gamma-ray spectra to headquarters. The AUTORAMP continued to operate flawlessly throughout the test period. Gamma-ray spectra were transmitted daily and analyzed at EML. The field test at McClellan AFB was extended to February 4, 1996.

Future RAMP research will focus on high speed data transmission and analysis. The use of INMARSAT-M satellites or INTERNET e-mail for two -way communications will provide EML with the ability to control or modify from NY all the operations of the RAMS at remote locations and to recover gamma-ray data instantaneously on demand.

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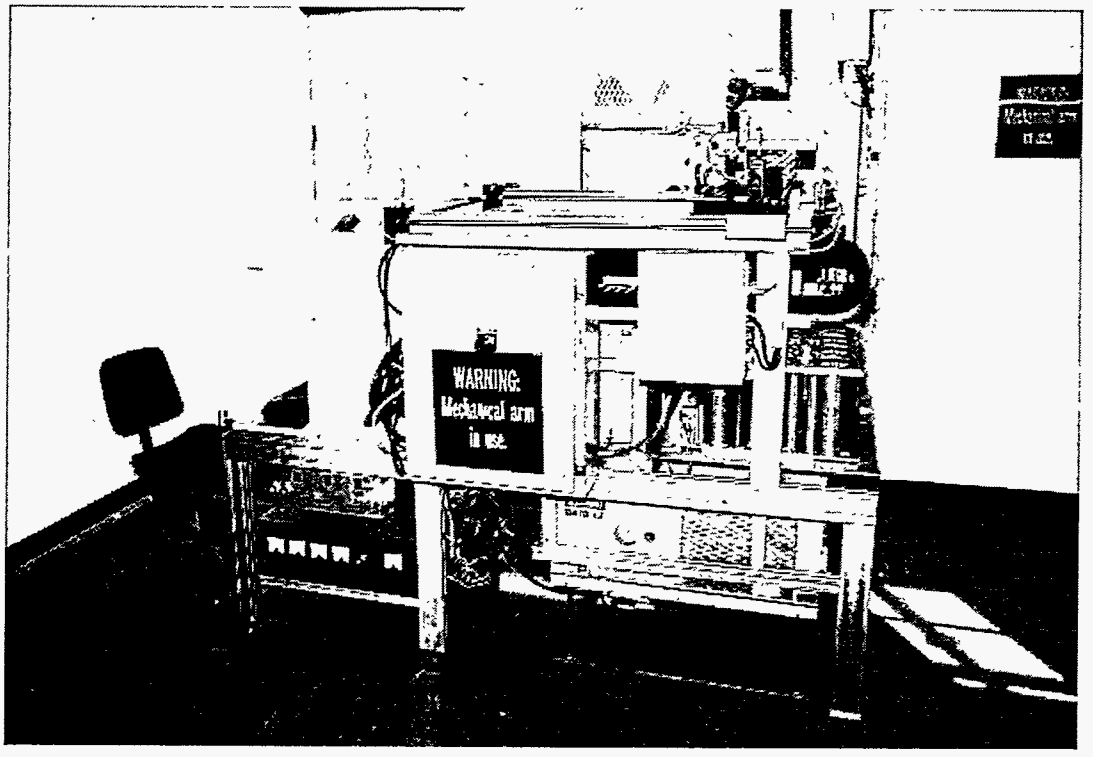

Figure 1.2 Automatic RAMP System.

1.5 MONITORING FOR ATMOSPHERIC RELEASE DURING CHINESE AND FRENCH UNDERGROUND NUCLEAR TESTS

Matthew A. Monetti, John Kada, Richard J. Larsen, Hsi-Na Lee, Colin G. Sanderson, Karin M. Decker, William Rivera, and Kevin J. Clancy

During 1995, both China and France conducted underground nuclear weapons detonations to test recent developments in their nuclear programs, although these tests were formally criticized by other nuclear nations as being unnecessary. The Chinese government conducted a single test on August 17, 1995 at their Lop Nor test site in north-western China. Between September 1995 and February 1996, France performed a series of six underground nuclear tests at its test sites at Mururoa and Fangataufa Atolls in the Pacific Ocean. In spite of reassurances given by the Chinese and French government, the tests evoked widespread expressions of public concern throughout the regions, as well as the world, in part because of fears that anthropogenic radioactivity would be released into the environment.

To help provide objective data relevant to these concerns, EML increased its monitoring of radionuclide activities in surface aerosol and precipitation at sampling sites it maintains under its Global Fallout, SASP, and RAMP programs (see Summary Nos. 1.2, 1.3 and 1.4). The sites utilized to investigate potential releases from the Chinese test were: Bangkok (14° N, 101° E), Chiang-Mai (19° N, 99° E), Taipei (25° N, 122° E), Seoul (37° N, 127° E) and Hong Kong (22° N, 114° E). Those involved in the Pacific region during the French tests were: Norfolk Island (29° S, 168° E), Wellington (41° S, 175° E), Easter Island (27° S, 109° W), American Samoa (14° S, 170° W), Lima, Peru (12° S, 77° W), and Chacaltaya, Bolivia (16° S, 68° W). Our sampling procedures were modified to increase the sampling frequency and to hasten the return and analysis of the samples. Additionally, forward trajectory analyses were performed following each test to predict the transport of radioactivity that may have been released.

Following the Chinese test, weekly deposition samples were collected at Bangkok and Chiang-Mai for four weeks, and at Taipei, Hong Kong and Seoul deposition samples were collected for the months of August and September. All the samples were sent to EML and analyzed as quickly as was possible. In all, 14 deposition samples were analyzed by gamma spectrometry to investigate possible radioactive releases following the Chinese test. During the period of French testing, all three of EML's global network programs were utilized. On-site gamma spectral analysis of weekly air filter samples at the Norfolk and Wellington sites was performed using EML's RAMP system. Air filter samples collected at Easter Island, American Samoa and the South American sites were sent to EML by express shipments and analyzed by gamma spectrometry. Deposition samples were collected at Easter Island, American Samoa and Chacaltaya at weekly intervals instead of the routine monthly sampling protocol. These samples were also quickly delivered to EML for gamma spectrometric analysis. The 164 samples analyzed for the French testing study include 44 RAMP samples, 69 SASP aerosol samples and 51 deposition samples.

Results of the gamma spectrometric analyses of these samples did not indicate that any radioactive material had been released to the atmosphere by the Chinese and French underground tests. If, in the unlikely event there was an atmospheric release of radioactivity, these results would

suggest that it was rather limited in quantity and/or distribution. These special studies have now been discontinued since no further testing is expected, and the sampling sites have resumed the normal operating procedures. The samples collected for these studies are being archived and will be available should any interest in follow-up studies develop.

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

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

The Environmental Measurements Laboratory has been identified as the Calibration Center for Radioactivity/Quality Assurance Science Activity Center (QA/SAC) for the World Meteorological Organization's (WMO) Global Atmospheric Watch (GAW) Program. In this role, EML will provide direction to GAW in measuring naturally-occurring radionuclides, specifically ^{7}Be, ^{210}Pb, ^{222}Rn and ^{222}Rn progeny. Quality assurance issues related to the sampling and analyses of these isotopes, such as sampler volume determination, filter paper efficiency, and gamma-ray spectrometric analyses, will be identified and guidance will be provided.

Philip W. Krey, Acting Director of EML, is a member of the Principals' Committee that oversees the QA effort for GAW. In October 1995, Mr. Krey gave a poster presentation on "EML's Global Network and Planned Role in GAW" at the First Dual Conference of the WMO/GAW and the International Global Atmospheric Chemistry (IGAC) on the Measurement and Assessment of Atmospheric Composition Change held in Beijing, China. During the conference, Dr. Ernst Brunke from the Atmospheric Trace Gas Research Station, Cape Point, South Africa, and Professor Zhongxiang Hong, Director of the Institute of Atmospheric Physics of the Chinese Academy of Sciences, each requested EML radon analyzers for installation at their measurement stations. Following the conference, Mr. Krey visited the newly established Chinese Global Atmospheric Watch Baseline Observatory (CGAWBO) at Mount Waliguan. During the visit, Dr. Tong Jie, Deputy Director of the Observatory, expressed interest in obtaining a Remote Atmospheric Measurements System (RAMS) from EML (see Summary No. 1.4). EML is currently evaluating the deployment of its radon and RAMS systems.

1.7 RADIONUCLIDES IN THE ARCTIC OCEAN

Thomas M. Beasley

During 1995, samples collected from the Trans-Arctic Ocean Section (TAOS) cruise of 1994 were analyzed for ^{129}I (water) and plutonium isotopes (sediment). In addition, ^{129}I was determined in water samples collected in 1986 from the Beaufort Sea near the coast of Alaska. The purpose of the analyses was to further characterize the distribution of these radionuclides in the Arctic Ocean as part of the Office of Naval Research's Arctic Nuclear Waste Assessment Program (ANWAP).

Iodine-129 in Arctic Ocean Waters. Figure 1.3 shows the results of the ^{129}I analyses from the TAOS collections. At each station, ^{129}I concentrations are maximal at shallow depths ($\leq$ 100 m); at none of the locations do ^{129}I concentrations reach those measured in samples collected from the Barents, Kara, and Laptev Seas during the period 1992-1993. They are more comparable to those measured in the Canadian Basin from collections made in 1993 and 1994 (Beasley, 1995).

In contrast to the TAOS samples, waters collected near the Alaskan Coast in October, 1986 (Beaufort Sea) show: (1) a subsurface maximum in ^{129}I between 300-400 m (Figure 1.4); and (2) elevated ^{129}I at the surface most probably due to the Chernobyl accident in late April of that year. The inventory of ^{129}I from 0-1500 m (3.7×10^{13} atoms m^{-2}) is estimated to be more than an order of magnitude lower than inventories, over comparable depths, in 1993-1994.

If ascribed exclusively to fuel reprocessing releases at Sellafield (U.K.), the 1986 inventory would, at most, double that derived from natural ^{129}I production and above-ground nuclear weapons tests. Because discharges from Sellafield have introduced considerable amounts of ^{137}Cs and other radionuclides to the Arctic Ocean (Cochran et al., 1995; Kershaw and Baxter, 1995 and references therein), ^{137}Cs measurements are in progress to confirm Sellafield as the source of the ^{129}I in these waters.

Plutonium Isotopes in Arctic Basin Sediments. The analysis of surface sediments collected on the TAOS cruise has confirmed the presence of low-ratio Pu throughout the Arctic Ocean basins (Beasley, 1995). Figure 1.5 shows collection sites and the measured $^{240}Pu/^{239}Pu$ sedimentary ratios. Where measurable, $^{241}Pu/^{239}Pu$ in these sediments indicate that in the basin interiors, the "age" of the Pu (i.e., its production age) is near 1957.

The absence of low-ratio Pu in the Greenland ice sheets between 1956 and 1965 (Koide et al., 1985) and the correspondence between the $^{240}Pu/^{239}Pu$ ratios measured there (0.17-0.28) with those measured in soils above 55^0 N in 1970 (0.18-0.19; Krey et al., 1976) argue against weapons-test debris as the source of this Pu. Sellafield discharges in the period 1958-1965 did contain Pu whose $^{240}Pu/^{239}Pu$ ratios ranged between 0.06 and 0.08 (Kershaw et al., 1995), but these ratios increased steadily through 1983 reaching values as high as 0.25. The presence of Sellafield-derived ^{129}I throughout much of the Arctic Ocean Basin would argue against the exclusive input and sedimentation of early Sellafield wastes (low-ratio Pu) to the exclusion of those released during the 1970s and 1980s (high-ratio Pu), particularly when transit times from the Irish Sea to the Barents and Kara Seas can be accomplished within a decade or less (Livingston, 1988; Smith et al., 1990). Thus, the origin of the low-ratio Pu in sediments as far west as the Canadian Basin suggests a more localized source in the Arctic itself.

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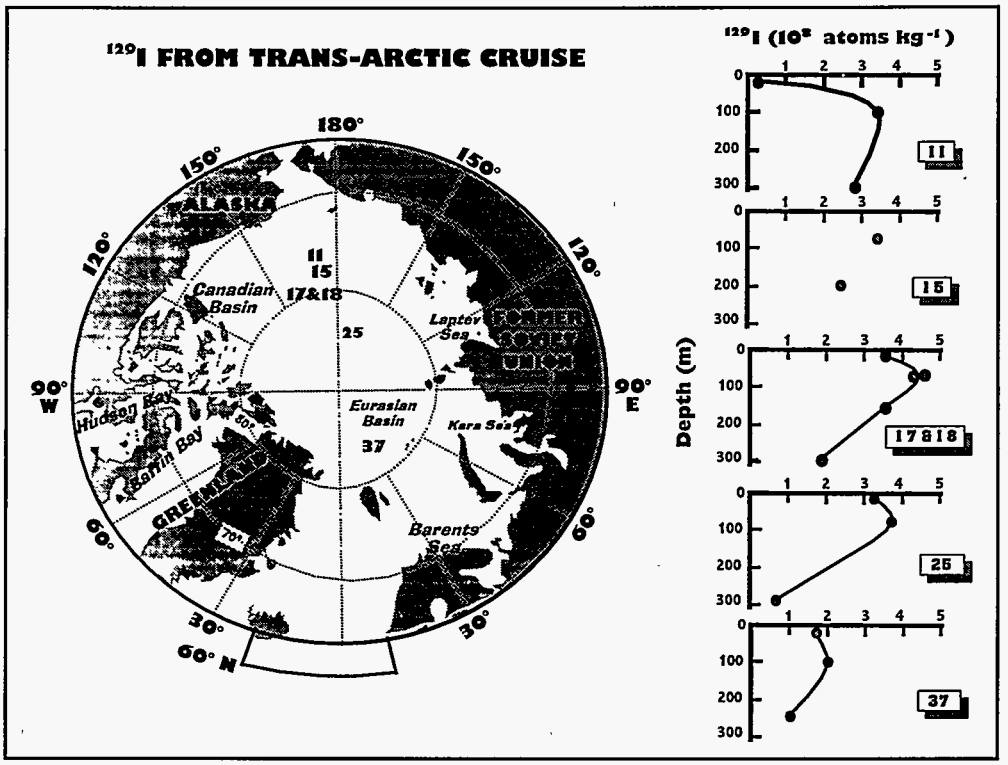

Figure 1.3 Trans-Arctic cruise sampling sites showing depth distributions of ^{129}I.

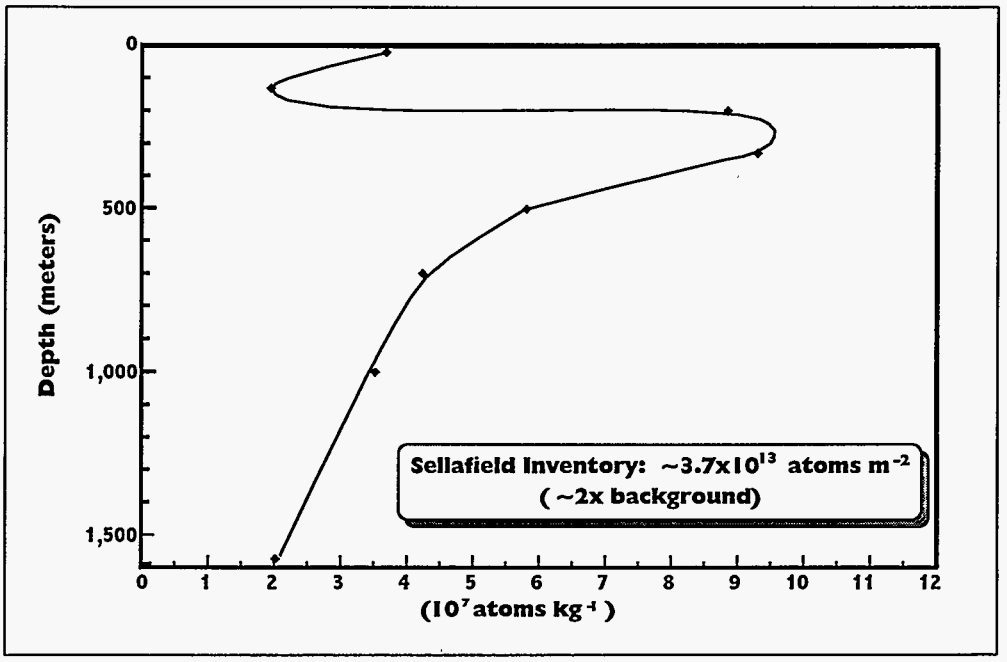

Figure 1.4 Profile of ^{129}I in waters from the Beaufort Sea collected in 1986. Elevated concentrations at the surface indicate input from Chernobyl accident.

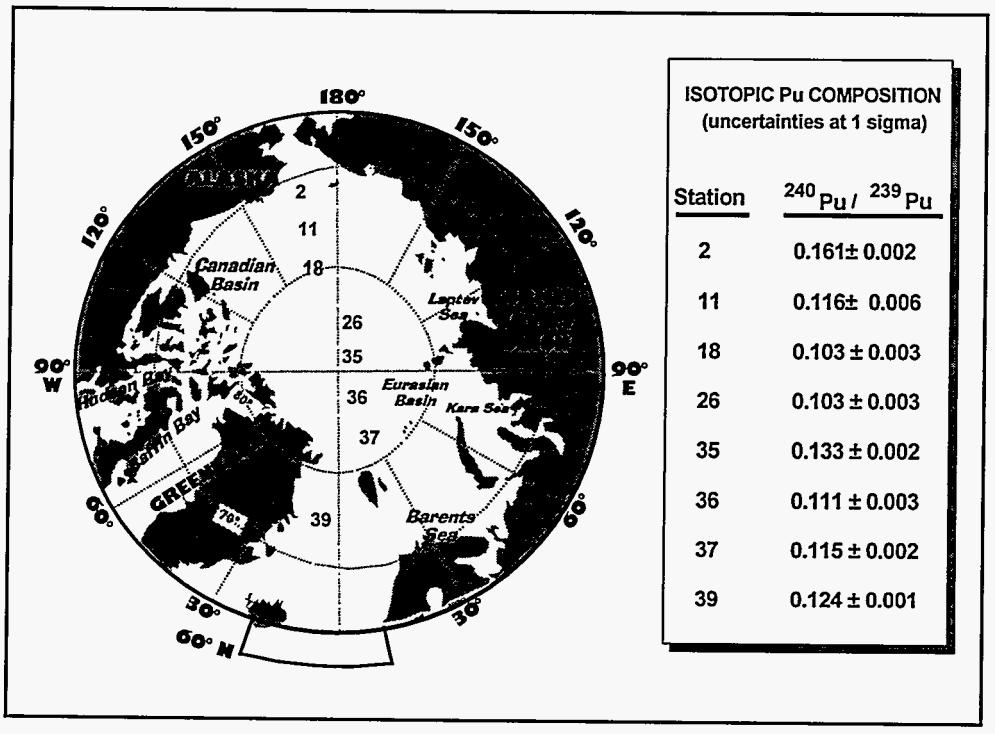

Figure 1.5 Plutonium isotope ratios in samples collected during Trans-Arctic Cruise showing wide spread distribution of low-ratio Pu throughout Arctic Ocean basins.

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

Thomas M. Beasley

During 1995, soil core collections at the Idaho National Engineering Laboratory (INEL) were completed as part of a study to determine the dispersion of Pu away from the Radioactive Waste Management Complex (RWMC). A total of 100 cores (0-10 cm depth) have been blended, tested for homogeneity, and analyzed for gamma-emitting radionuclides. Approximately 30 cores were taken outside of the INEL site boundaries; the remaining cores were collected at various distances from the RWMC (Beasley, 1995). Subsamples of all cores have been processed for mass spectrometry measurements of Pu isotopes and ^{237}Np; approximately 50% of the analyses has now been completed.

Figure 1.6 shows representative ^{240}Pu/^{239}Pu ratios for cores collected in the direction of the principal winds at the site. Global fallout values for the ^{240}Pu/^{239}Pu ratio at the INEL are expected to lie within the range determined by Krey et al. (1976), namely 0.176 ± 0.014. From the data shown in Figure 1.6, it is clear that substantially lower ratios are observed in several of the cores collected southwest of the RWMC. For example, assigning a ^{240}Pu/^{239}Pu ratio of 0.05 to RWMC Pu (a value typical of Pu processed at Rocky Flats, CO) the amount of RWMC-derived Pu at coring site 56 is estimated to be ~ 40 percent of the measured 70 ± 1 Bq m^{-2} of total ^{239}Pu + ^{240}Pu at that location.

Because winds can reach velocities > 100 km hr^{-1} at the INEL, redistribution of surface soils is constantly taking place. Consequently, Pu released from the RWMC in the late 1960s and early 1970s can be transported to great distances. Equally probable, however, is that soils once contaminated with RWMC-derived Pu can be winnowed over time and replaced or mixed with soil carrying only a fallout Pu signature. It is not surprising, therefore, that in the intervening three decades between Pu release at the RWMC and our soil sampling, redistribution processes have operated to blur what may have initially been a more easily identifiable "footprint" of the Pu released at that facility.

The Idaho Chemical Processing Plant (ICPP) has processed highly-enriched uranium (HEU) over its operating history (maximum percent ^{235}U = 100; Benedict et al., 1981). Thermal neutron irradiation of ^{235}U produces ^{236}U and ^{237}Np. We analyzed core 54, collected near the ICPP, for ^{236}U and find atom concentrations of (7.8 ± 1.1) x 10^{8} atoms g^{-1} soil and a ^{235}U/^{238} atom ratio of 0.007296 ± 0.000011, a value that exceeds that of natural uranium, i.e., 0.00725. Because ^{236}U has not been reported as measurable in global fallout, its presence at site 54 must result from stack emissions at the ICPP. Moreover, the concentration of ^{237}Np at site 54 (~ 2 x 10^{8} atoms g^{-1} soil) is elevated (30-40%) with respect to those we have measured in other cores to date.

We have confirmed the presence of both ^{236}U and ^{237}Np in the ICPP waste stream by analyzing water samples from monitoring wells in the Snake River Plain aquifer downgradient from this facility. Near the ICPP, ^{236}U and ^{237}Np concentrations were 3 x 10 ^{11} and 1 x 10 ^{9} atoms 1 $^{-1}$, respectively. Uranium-236 could be traced to distances of ~10 km while ^{237}Np was measurable only

within 2 km of the plant. We plan further core analyses for ^{236}U to determine the extent to which it can be detected across the INEL site.

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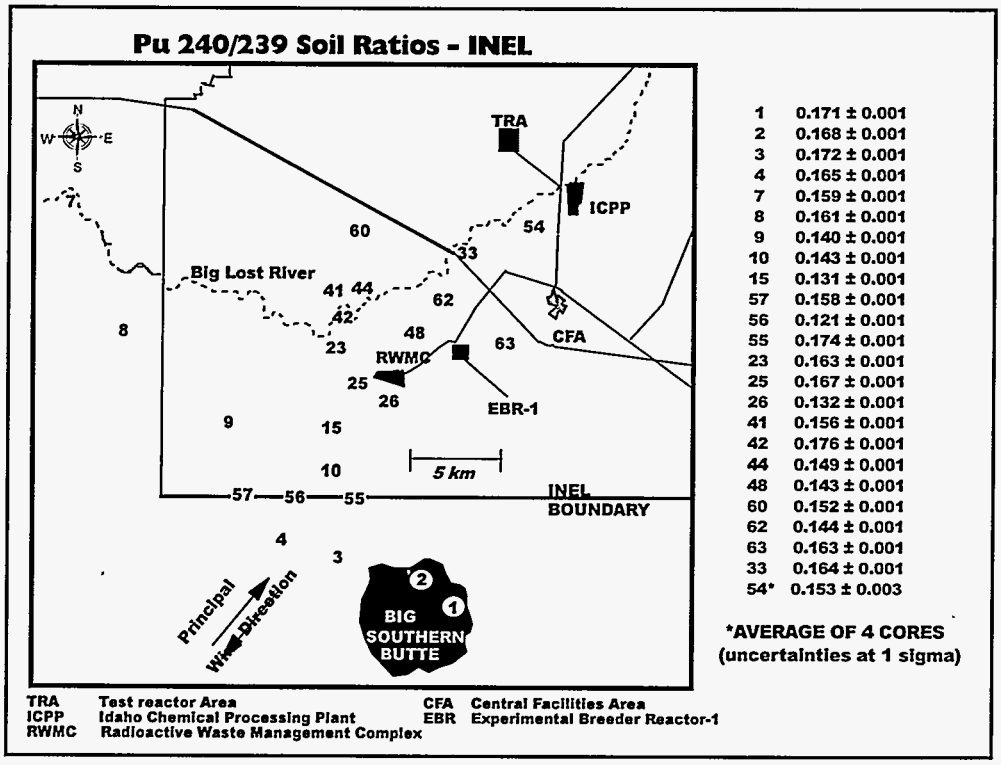

Figure 1.6 Typical $^{240}Pu/^{239}Pu$ ratios in soils at the Idaho National Engineering Laboratory.

1.9 HISTORY OF DIOXIN-LIKE COMPOUNDS IN LAKE SEDIMENT CORES

Matthew A. Monetti, John Kada and Merrill Heit

For this study, EML has collaborated with the EPA's National Center for Environmental Assessment in Washington, D.C., to examine the chronological record of deposition of dioxin-like compounds, i.e., dioxins, furans and coplanar polychlorinated biphenyls (PCBs), revealed in sediments from lakes located in the contiguous United States and Alaska. Since these compounds are highly toxic and are likely to be human carcinogens, the EPA, as part of their Dioxin Exposure Initiative, has an interest in determining the temporal trends in the levels, patterns and profiles of dioxins, furans and PCBs. EML has been involved with these types of investigations since the 1970s. Over the past twenty years, EML has collected sediment cores from 64 lakes or reservoirs in the U.S. and an archive of these samples has been maintained at the Laboratory.

Sediment cores from 11 lakes (Figure 1.7) were chosen for investigation in this study based on certain criteria, including distance from known sources of dioxin-like compounds and lack of point-sources of dioxin-like compounds in the watershed and regional representative lakes having good sediment-dating resolution in the time-period of interest (back to 1930 or further). As a result of previous studies, data was previously available for ^{137}Cs and, in some instances, excess ^{210}Pb, which was used to establish the sedimentation rates and chronology for the cores. Samples representing ten year intervals were prepared from the cores. Some samples, particularly those from pre-industrial sections of the core, represented larger time periods. Eighty samples (including three blind replicates) were analyzed for dioxins, furans and PCBs by a contractor laboratory selected by the EPA. The contractor followed EPA Method 1613 which uses high resolution gas chromatography / high resolution mass spectrometry techniques. Results included concentrations of total congener group for tetra through octa-chlorinated dioxins and furans, in addition to several 2,3,7,8 -substituted isomers and several PCBs (77, 118, 105, 126, 156 and 169). The database is likely to be the largest of its kind, containing 2,464 values for dioxin-like compounds.

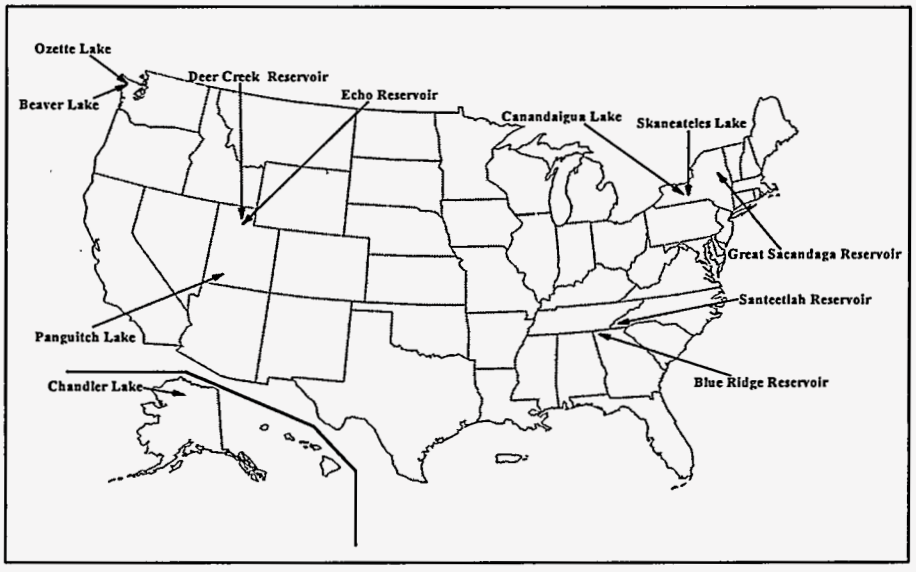

Figure 1.7 Sediment core sample locations.

1.10 ENVIRONMENTAL RADIATION MEASUREMENTS USING PRESSURIZED IONIZATION CHAMBERS (PICs)

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 continued at our regional baseline station in Chester, NJ. Additional monitoring is done at Princeton, NJ. This study has been part of a long-term program whose goal is to document the natural variations as well as any anthropogenic perturbations in the natural background.

A comprehensive analysis of the eighteen year record of daily average dose rates at our Chester station was completed this year. The focus of the study was a quantitative description of the temporal variations that result primarily from snow cover, soil moisture, and atmospheric scavenging of radon progeny. Figure 1.8 is a plot of the eighteen year record illustrating the overall magnitude and daily variations in the dose rate. These results were summarized in a paper presented at the Natural Radiation Environment VI Symposium, Montreal, Canada (Shebell and Miller, in press). The paper also provided means and extremes of the entire record. An analysis of the data by season indicated that the winter months exhibited the largest variation and lowest dose rate. Frequency histograms were generated to show the variations in the daily average dose rate.

The analysis and summary of the eighteen year database provided a natural conclusion to routine monitoring at Chester. A decline in, and a reallocation of resources, led to the decision to terminate the program at Princeton as well. Regional environmental monitoring with PICs will probably continue in a limited capacity.

Monitoring began at Brookhaven National Laboratory (BNL) as part of the 11th International Intercomparison of Environmental Dosimeters. The initial 42-day record of one-minute exposure rates showed a brief but dramatic increase in the exposure rate at noon on the 10th of December. The elevation lasted for only two consecutive one minute measurements and was about six times the 42-day average. It has not been linked to any activities or known releases at BNL. The possibility of equipment malfunction has not been eliminated. An additional system has been deployed to further investigate the situation, as well as to provide a backup unit.

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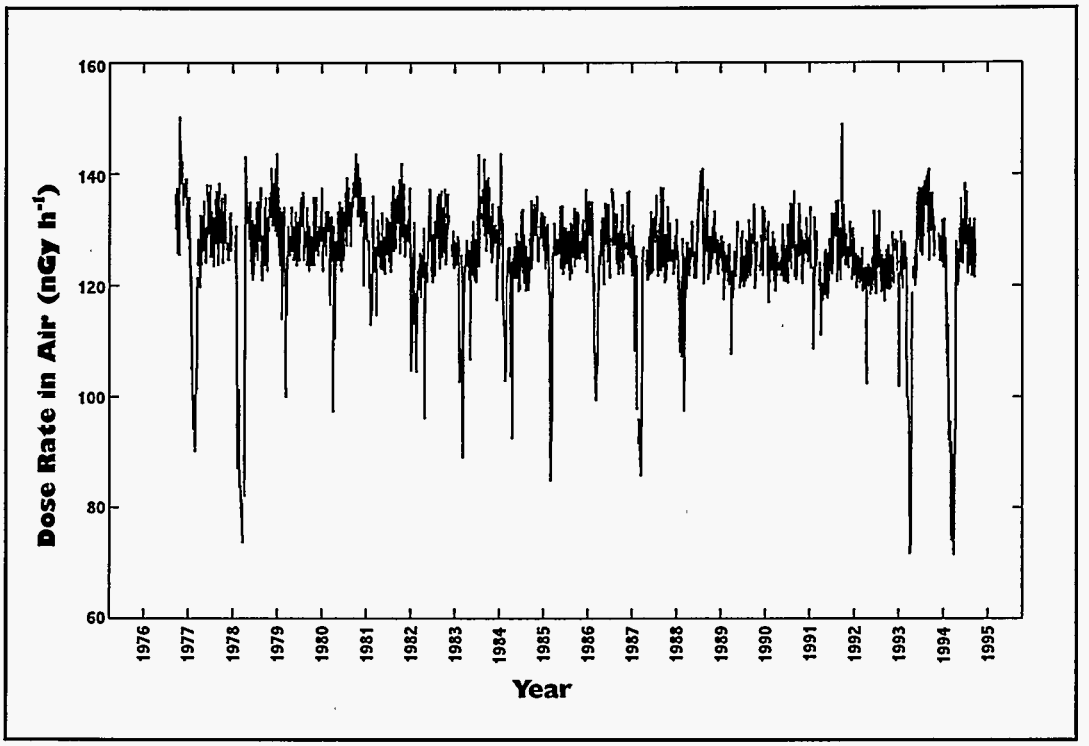

Figure 1.8 Average daily dose rate in air at EML's regional baseline station in Chester, NJ, from 9/1/76 - 9/1/94.

1.11 ENVIRONMENTAL THERMOLUMINESCENCE DOSIMETRY

Gladys A. Klemic and Nestor Azziz

EML's thermoluminescence dosimetry (TLD) program includes maintaining state-of-the-art instrumentation for routine environmental radiation monitoring, the planning and implementation of international intercomparisons, contributions to the development of national standards, special field applications (see Summary Nos., 2.6, 2.7 and 6.18) and investigations of the use of TLDs in mixed gamma-neutron radiation fields (see Summary No. 2.8).

EML maintains two TLD readers in addition to 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 for the international intercomparisons and batch testing of chips. A commercially available manually operated, linearly heated planchet reader is being used for research applications, including glow curve analysis. In 1995, EML's ^{137}Cs calibration facility was upgraded to include improved sample positioning and the use of recalibrated Shonka-Wyckoff chambers to provide traceability to the National Institute of Standards and Technology (NIST).

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 are presently the only available large scale and universally recognized quality assurance program for passive environmental dosimetry. Participation in the program is voluntary and results are reported without identifying individual participants.

Invitations for the 11th Intercomparison were mailed in late 1995, and 125 participants from 36 countries have registered to participate. The intercomparison will be held at BNL in collaboration with NIST and Idaho State University. An undisturbed field at BNL was characterized by *in situ* gamma spectrometry and ionization chamber measurements and was selected for the site of field deployment. Routine monitoring with EML's pressurized ionization chamber (see Summary No. 1.10) and TLDs began in December 1995.

National Standards Work

In April 1995, EML hosted a meeting of the American National Standards Institute (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 National Voluntary Laboratory Accreditation Program (NVLAP). The draft of ANSI N13.29 was submitted to the Health Physics Society in late 1995 and is presently under review. Results from the International Intercomparisons of Environmental Dosimeters have proved useful in helping to set procedures and tolerance levels for these standards.

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 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 are an important part of this program. Using such codes, calculations are performed to determine radiation fields from instrument readings, to relate measured fields to the properties and distributions of radiation sources, and to 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 concentrated on neutron dosimetry because of the continuing difficulties associated with making reliable assessments of neutron fields and exposures. Highly conservative estimates of neutron dose equivalents, which must be used when measurements are uncertain, affect the efficiency and cost of DOE facility operations. Because of the experience gained and expertise developed at EML in this field, notably with multisphere neutron spectrometer (MNS) 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. The long-term measurements at the Princeton Tokamak Fusion Test Reactor (TFTR), described in Summary No. 2.2, have helped assure that the TFTR shielding would be adequate for high-power operation and will contribute to accurate shielding calculations for the next generation of fusion reactors. Neutron spectrum measurements at the Army Pulse Radiation Facility (APRF) (see Summary No. 2.3) were performed at the request of the Defense Nuclear Agency. This study 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. In support of our neutron spectrometry measurements, we have changed and clarified our methods of unfolding neutron spectra from MNS data (see Summary No. 2.4) and are calculating the energy response of our new high-sensitivity MNS (see Summary No. 2.5).

There is increasing awareness that aircraft crews are exposed to secondary cosmic radiation and receive one of the highest average dose equivalents of any occupationally exposed group. However, the atmospheric ionizing radiation field is highly complex, and there is significant uncertainty in our knowledge of the high-energy neutron component, which can cause up to half or more of the biological damage. 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. These measurements are described in Summary No. 2.6. 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 response to concerns about radiation safety for such aircraft, NASA's Langley Research Center (LaRC) and EML have planned a series of high-altitude measurements of atmospheric ionizing radiation (AIR) using the NASA ER-2 (enhanced U-2) aircraft. The primary instruments will be the ones EML used on the Canadian flights, but a number of other radiation measurement groups will also participate. The AIR project is described in Summary No. 2.7.

Dosimetry systems which are *not* sensitive to neutrons are also important for measurements in complex radiation fields. A study to improve our understanding of the response to neutrons of various thermoluminescent dosimeter (TLD) materials primarily sensitive to low-LET radiation is described in Summary No. 2.8.

Although this program primarily involves research in support of radiation protection activities, it also provides 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 and other government agencies, 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 1995, have been the Princeton Plasma Physics Laboratory, APRF, Science Applications International Corporation, NASA LaRC, the University of Akron, Los Alamos National Laboratory, the Royal Military College of Canada, and the Defense Research Establishment, Ottawa. For the AIR project, new collaborations have been initiated with the German Air and Space Research Institute (DLR) and Kiel University, the National Radiation Protection Board of Britain, the University of San Francisco, the Boeing Company, the NASA Johnson Space Center, and Apfel Enterprises.

2.2 MEASUREMENT OF LEAKAGE NEUTRON SPECTRA OUTSIDE THE SHIELDING OF THE TOKAMAK FUSION TEST REACTOR

Paul Goldhagen, Marcel Reginatto, Peter Shebell,
William Van Steveninck, and Alfred J. Cavallo

The Tokamak Fusion Test Reactor (TFTR) of the Princeton Plasma Physics Laboratory

(PPPL) has been the flagship of America's magnetic-confinement fusion program. In 1994 and 1995, it produced record-breaking pulses of fusion power (>10 MW) using the deuterium-tritium (D-T) reaction. 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.

The EML measurements were 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 yr^{-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.

Neutron measurements were performed using a multisphere neutron spectrometer (MNS) with 12 detectors operating simultaneously. The spectrometer incorporated computer-controlled gated electronics to allow analysis of individual TFTR fusion pulses (~1 second duration, 7 to 15 minutes apart). In addition, data were collected during the time between TFTR pulses and recorded separately to measure the spectrum of background cosmic-ray neutrons. Measurements were performed 1 m outside the north wall of the Test Cell (about 20 m from the center of TFTR) and 120 m north-northeast of the center of TFTR in a trailer near the site boundary fence. Gamma-ray measurements were also made at these and other locations (Kugel, et al., 1995). Data from more than 8,000 TFTR pulses have been recorded, which is more than enough to determine neutron spectra near the shielding for D-D fusion and at both locations for D-T fusion, but we are continuing our measurements at the 120 m location 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.

Normalized neutron energy spectra (fraction of fluence as a function of energy) were unfolded from the MNS measurements as described in Summary No. 2.4 and are shown in Figure 2.1. As is customary for neutron spectra displayed on a logarithmic energy scale, the fluence per lethargy is plotted, where lethargy is the natural logarithm of the energy. On such a plot, the typical moderated neutron spectrum (fluence per energy proportional to 1/E) is a horizontal line and equal areas represent equal numbers of neutrons. As expected, the neutrons escaping from the shielding for D-T fusion have higher energies than for D-D fusion, but neither spectrum shows a significant number of unmoderated fusion neutrons (14.1 MeV for D-T, 2.5 MeV for D-D). The ambient dose equivalent, H^*_{10}, per unit fluence calculated for the D-T spectrum is 2.5 times that for the D-D spectrum. Absolute fluences and dose equivalents per TFTR fusion neutron will be determined when the D-D and D-T neutron yield of each TFTR pulse is determined, sorted and summed.

Results of this work were presented at the Fourth Annual Meeting of the Council on Ionizing Radiation Measurements and Standards, November 28-30, 1995, and at the American Nuclear Society Topical Meeting "Advancements and Applications in Radiation Protection and Shielding", April 21-25, 1996, in Falmouth, MA.

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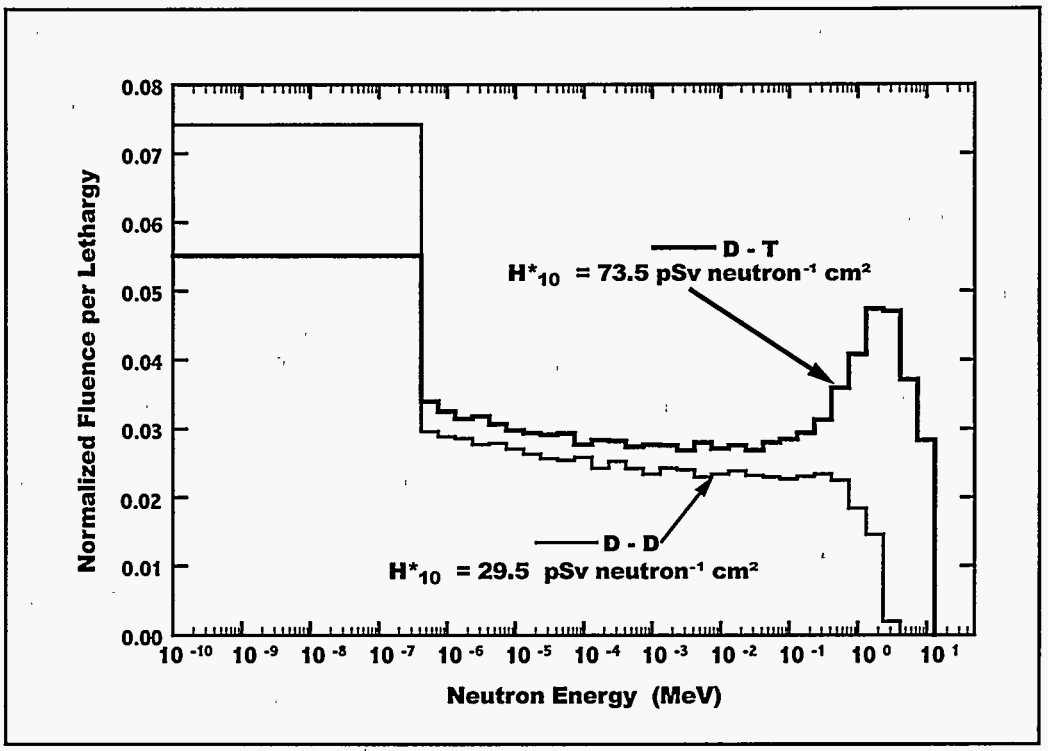

Figure 2.1 Neutron spectra measured 1 m outside the north wall of the TFTR Test Cell during operation with D-D and D-T fusion. With neutron energy plotted on a logarithmic scale, the spectra are plotted as fluence per lethargy ($\ln(E_n)$) so that equal areas represent equal fractions of the neutron fluence. H* is the ambient does equivalent.

2.3 NEUTRON SPECTRUM MEASUREMENTS AT DISTANCES UP TO 2 KM FROM A FISSION SOURCE AT THE U.S. ARMY PULSE RADIATION FACILITY

Paul Goldhagen, Marcel Reginatto, Peter Shebell,
Gladys A. 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 Aberdeen Proving Ground 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 et al., 1994, 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 (MNS) 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. In 1993, the distances were 300, 1588, and 1986 m. Neutron spectra were unfolded from the background-subtracted counts in each detector of the MNS (see Summary No. 2.4 for the measured and calculated spectra).

It is difficult to quantitatively compare the calculated spectra with the unfolded measured spectra, so we have made a more direct comparison. We have folded the calculated spectra with our detector response functions, i.e., numerically integrated their product over neutron energy, and compared the results with the measured counts in each detector. The ratios of the calculated to measured (c/m) counts in each detector are shown in Figure 2.2, plotted against range in meters.

In spite of the spread in the c/m count ratios shown in Figure 2.2 (0.65 to 1.68 at the extremes), the agreement is good, especially compared to the factors of 5 to 10 in the c/m activation ratios at Hiroshima. Furthermore, most of the differences at APRF can be understood by taking into account the difference between the environmental conditions (air density and humidity and ground moisture content) during the measurements and those assumed for the calculation. The high c/m ratio at 715 m can be explained by the presence of trees blocking roughly 20-40% of the sky (and skyshine neutrons) at that site.

We conclude that the Hiroshima neutron discrepancy is probably <u>not</u> primarily due to problems with the long-range transport calculations. We also note that the falloff of the thermal neutron fluence with distance we measured at APRF closely matches that for the thermal neutrons at Hiroshima determined from activation measurements. This suggests that many more nearly

unmoderated fission neutrons (or at least neutrons above the oxygen cross section window at 2.35 MeV) escaped from the Hiroshima weapon than were assumed in the DS-86 model. 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 all types of radiation would be affected.

Results of this work were presented at the Fourth Annual Meeting of the Council on Ionizing Radiation Measurements and Standards (November 28-30,1995), and at the American Nuclear Society Topical Meeting "Advancements and Applications in Radiation Protection and Shielding", April 21-25, 1996, in Falmouth, MA, and will be published in its Proceedings.

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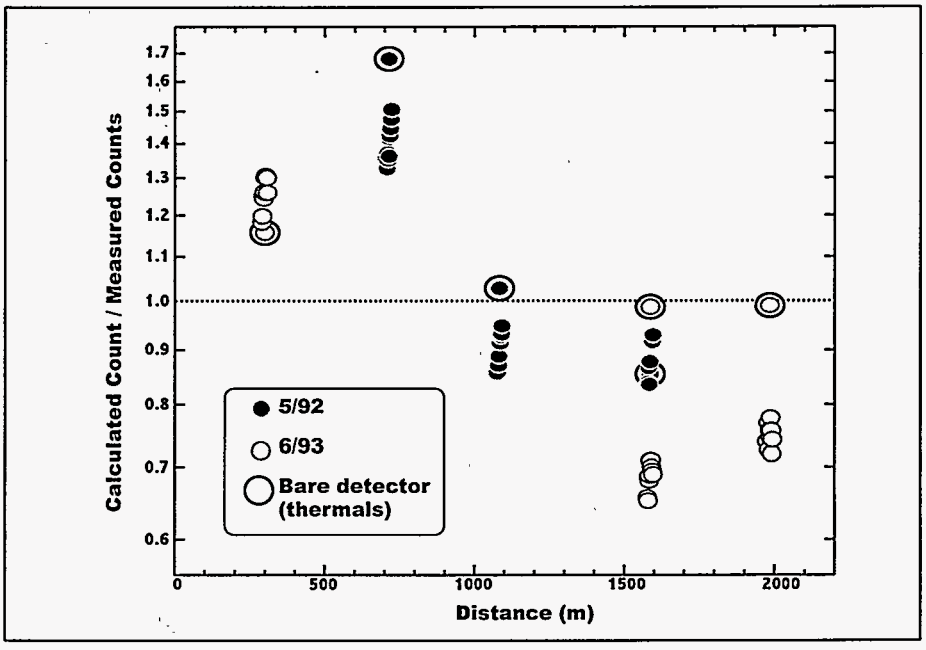

Figure 2.2 Ratio of calculated to measured neutron counts in each sphere detector vs. distance from APRF critical assembly. Calculated counts were computed by folding Kaul's calculated neutron spectra with EML response functions. At each location, smaller spheres (low neutron energy) are plotted to the left of larger spheres (high neutron energy) to produce a slant if there is a trend with neutron energy.

2.4 DECONVOLUTION OF EML MULTISPHERE NEUTRON SPECTROMETER DATA

Marcel Reginatto and Paul Goldhagen

The set of detectors in a multisphere neutron spectrometer (MNS) consists of counters sensitive to slow neutrons which are surrounded by spherical polyethylene moderators of different sizes to produce detectors with different energy responses. For a given detector, the number of counts N is related to the response function R(E) and the neutron fluence f(E) by an integral over the energy E of the form

$$N = \int R(E)f(E)dE .$$

Deconvolution (unfolding) of the data amounts to finding a spectrum f(E) that fits the data for all detectors. The problem of reconstructing the neutron spectrum from a finite number of measurements is mathematically ill posed, and the solution is not unique. Therefore, to find a physically relevant spectrum, *a priori* information must be implemented in the solution method.

A number of mathematical methods have been developed for the deconvolution of neutron spectra. One of the most commonly used unfolding programs is the SAND-II code package (McElroy et al., 1967; Berg, 1968). The calculational procedure used consists of the selection of an initial approximation spectrum (which incorporates *a priori* information), and subsequent perturbation of that spectrum by iterative adjustments to yield a solution spectrum that fits the data.

A version of the neutron spectra unfolding code SAND-II (RSIC code package CCC-112) which runs on personal computers was obtained from the Radiation Shielding Information Center and modified at EML to incorporate the EML MNS detector response functions. This modified code was then used for the deconvolution of data from measurements that EML carried out at the Princeton Tokamak Fusion Test Reactor (TFTR) and at the U.S. Army Pulse Radiation Facility (APRF).

Measurements of leakage neutron spectra were made outside the TFTR Test Cell during deuterium-deuterium (D-D) and deuterium-tritium (D-T) operation (see Summary No. 2.2). After examining spectra from tokamak shielding calculations, we used initial approximation spectra as follows: for D-D fusion, fluence per unit energy proportional to 1/E (constant fluence per unit lethargy) from 0.4 eV to 0.5 MeV then a sharper decrease up to a cutoff at 2.5 MeV; for D-T fusion, fluence per unit energy proportional to 1/E from 0.4 eV to 2.5 MeV then a sharper decrease up to a cutoff at 14.1 MeV. Figure 2.1 shows the results for the unfolded neutron spectra measured 1 meter outside the north wall of the TFTR Test Cell.

The measurements at APRF (see Summary No. 2.3) were made to test long-range neutron transport calculations using codes similar to those applied to the Japanese A-bomb survivor dosimetry. We, therefore, used spectra calculated by the code to be tested as the initial approximation spectra for unfolding measured spectra. The long-range transport calculation we tested was performed by Kaul (1995) using the discrete ordinates transport code DORT. Measured

and calculated APRF neutron fluence spectra for the five distances where we made measurements are shown in Figure 2.3 The fluence per lethargy (per joule of fission) has been multiplied by the square of the range to remove the $1/R^2$ dependence. The Kaul calculated spectra show considerable structure above 100 keV which increases with distance, reflecting the nuclear resonance structure of the oxygen and nitrogen in the air. Such structure appears in the unfolded spectra from SAND II when the calculated spectra are used as initial guesses. Since the resolution of our MNS is no better than 2 to 4 bins per energy decade, the fine-binned output was collected into 4 bins per energy decade, and these wide-binned measured spectra are shown in Figure 2.3. Since our MNS cannot resolve any spectral shape within the thermal-energy region, we have shown no measured thermal spectra. There was generally good agreement between our measured neutron spectra and those from the long-range transport calculation (see Summary No. 2.3).

References

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"Neutron and Gamma Ray Fluences Calculated for the APRF Reactor at Aberdeen, MD", private communication via electronic mail, August 15, 1995

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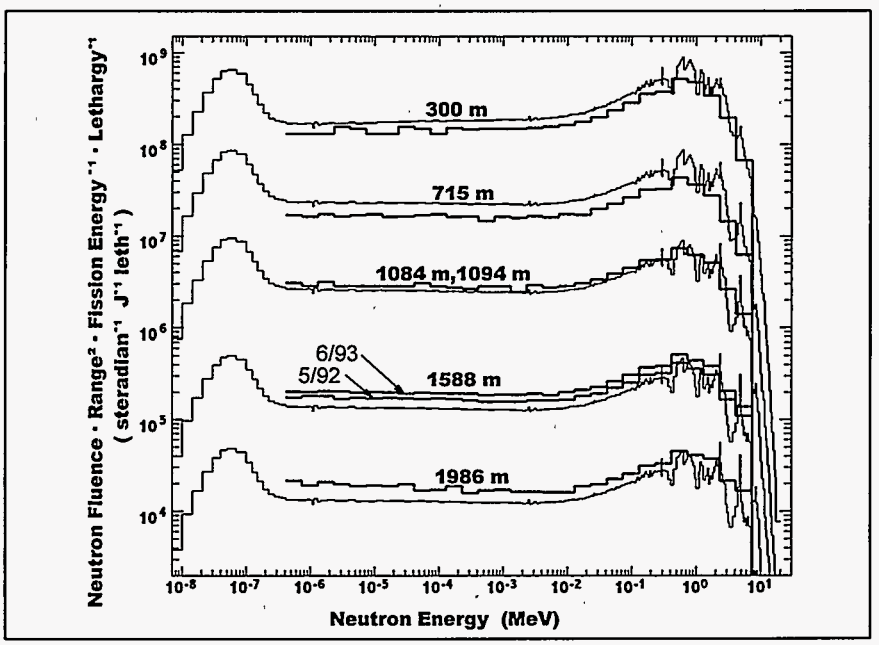

Figure 2.3 Measured and calculated APRF neutron fluence spectra. Coarse-binned spectra are unfolded EML measurements. Fine-binned spectra are calculations by Kaul.

2.5 CALCULATION OF MULTISPHERE NEUTRON SPECTROMETER RESPONSE FUNCTIONS

Paul Goldhagen

EML's primary instrument for measuring neutron energy distributions is the multisphere neutron spectrometer (MNS), which is a series of spherical polyethylene moderators of different sizes surrounding counters sensitive to slow neutrons. 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. The slow-neutron counters of the MNS used for the measurements at TFTR and APRF are pulse ionization chambers filled with $^{10}BF_3$ gas at a pressure of about 0.46×10^5 Pa. In 1994 we calculated the response functions of each of the 12 detectors of this MNS using the Monte Carlo particle transport code MCNP (Briesmeister, 1993) and the latest neutron interaction cross sections then available, ENDF/B-V (Goldhagen, 1995). In 1995, the new ENDF/B-VI set of neutron cross sections became available for use with MCNP. We have repeated the response function calculations for detector #1 (the bare $^{10}BF_3$ counter) and detector #6 (100.5 mm diameter, 0.4378 kg moderator) with the new cross sections, and there is no significant difference in the results. We plan to do more calculations with the new cross sections for the detectors with larger moderators.

For measurements of cosmic-ray neutrons on aircraft (see Summary Nos. 2.6 and 2.7), we are using a new MNS (Goldhagen and Van Steveninck, 1995) which has proportional counters filled with ^3He at a pressure of 4.0×10^5 Pa as its slow-neutron counters. We have now started to calculate the response functions for the new MNS. In collaboration with Timothy Kniss of the University of Akron, the response of all of the detectors will be calculated up to 20 MeV using MCNP. Figure 2.4 shows the response of a bare ^3He counter and the bare $^{10}BF_3$ counter. Also shown is the Maxwellian thermal energy distribution for 20 °C, which is the energy region of most importance. The response of the ^3He counter integrated over the thermal distribution is about 6.2 times the thermal response of the $^{10}BF_3$ counter. Neutrons from cosmic rays have energies up to 10 GeV and beyond. To calculate the response of the new MNS at energies above 20 MeV, we will use the Los Alamos High Energy Transport Code (LAHET) in collaboration with scientists at LANL and elsewhere.

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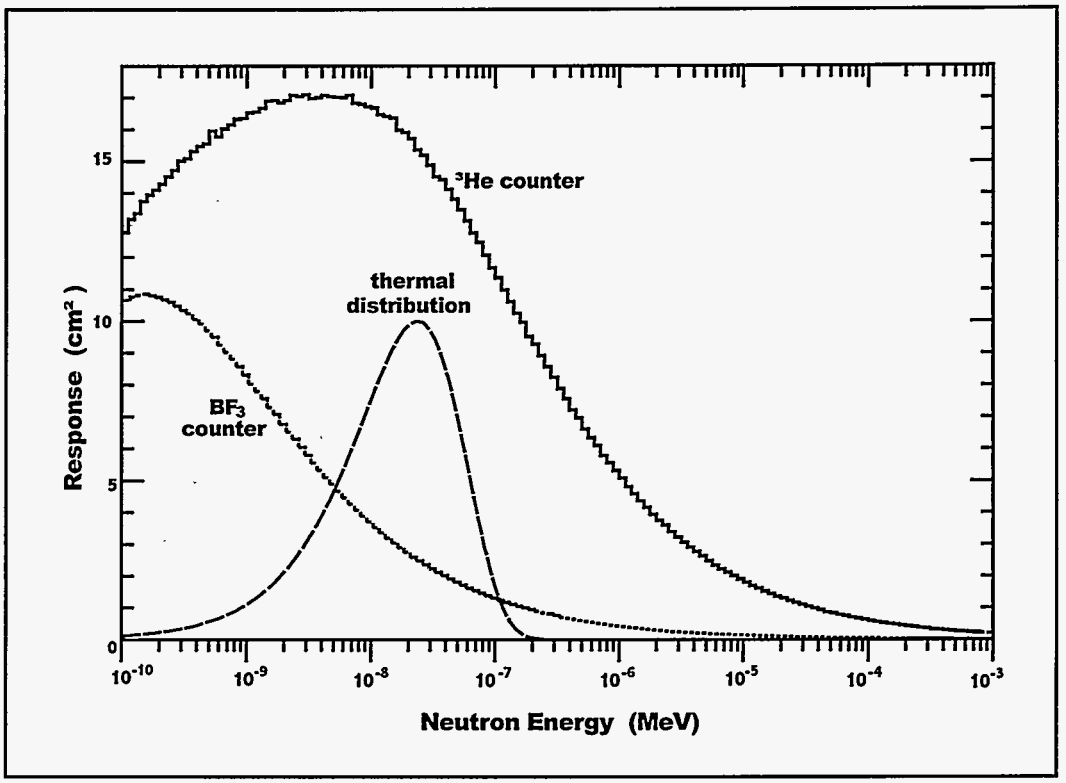

Figure 2.4 Calculated response functions for a bare ^{3}He counter of the new EML multisphere spectrometer and the bare $^{10}BF_3$ counter of the older spectrometer with $^{10}BF_3$ counters. The vertical axis units of effective area are equivalent to counts cm^2 $neutron^{-1}$. The thermal energy distribution for 20°C (in arbitrary units) is also shown.

2.6 MEASUREMENT OF COSMIC RADIATION ABOARD A CANADIAN FORCES JET AIRPLANE IN FLIGHT

Paul Goldhagen, William Van Steveninck, Alfred J. Cavallo and Peter Shebell

Crews working on jet aircraft are exposed to secondary cosmic radiation and receive one of the highest average dose equivalents of any occupationally exposed group in the western world. In spite of this, the atmospheric ionizing radiation field and dose equivalent rates from it are not precisely known (see Summary No. 2.7). In part, this uncertainty is due to the complexity of the field, which contains virtually every kind of radiation at all energies up to 10 GeV and beyond.

EML has joined 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 which is the component of the atmospheric radiation field that contributes the greatest uncertainty to the dose equivalent and to estimates of health risk. A successful test flight was made over Canada in December 1994, and transatlantic flights from Trenton, Ontario, to Cologne, Germany, and back were made on May 9 and 11, 1995, with the altitude varied in steps from 10.1 to 12.5 km (33,000 to 41,000 ft).

DREO and RMC flew a variety of radiation detectors. EML's detectors included our new ^{3}He-counter multisphere neutron spectrometer (MNS), a pressurized argon ionization chamber (PIC), thermoluminescent dosimeters (TLDs), 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 MNS results for response to high-energy protons.

The dose rate in air from charged particles and gamma rays measured by the PIC on the transatlantic flights is shown in Figure 2.5 plotted as a function of time. On the return flight, the increasing ionization dose rate as the geomagnetic latitude varied from 53° to 57° N while the altitude was constant at 37,000 and 39,000 ft shows the effect of decreasing geomagnetic shielding. The nearly constant ionization dose rate at constant altitude on the flight to Cologne while the geomagnetic latitude varied from 60° to 70° N shows that there is no further dose rate increase above 60° NGM. This saturation occurs because the earth is shielded from low-energy galactic cosmic rays by the magnetic field of the solar wind plasma that fills the solar system. The total PIC dose in air for the round trip was 34 ± 5 μGy. (The error will be reduced after calculating the effect of the PIC wall for the electron component as a function of altitude.) The dose measured by our Al_2O_3:C TLDs was 32 ± 6 μGy. Both of these instruments are relatively insensitive to neutrons.

MNS measurements with thousands of counts in each detector were recorded every 150 seconds on the transatlantic flights. At high geomagnetic latitude, the counts in each of the MNS detectors except the one responding to the highest energies were all 1.61 ± .05 times higher at 41,000 ft, than at 33,000 ft. This means that the shape of the neutron spectrum below 10 MeV did not change significantly with altitude in this range, while the dose equivalent from that portion of the neutron spectrum increased by a factor of 1.61. Neutron spectra and dose-equivalent rates for each altitude and geomagnetic latitude will be determined when we have completed calculations of the response functions of the MNS (see Summary No. 2.5) and the scintillation counters.

Preliminary results of this work were presented at the Fourth Annual Meeting of the Council on Ionizing Radiation Measurements and Standards (November 28-30, 1995), and at the American Nuclear Society Topical Meeting "Advancements and Applications in Radiation Protection and Shielding", April 21-25, 1996, in Falmouth, MA, and will be published in its Proceedings.

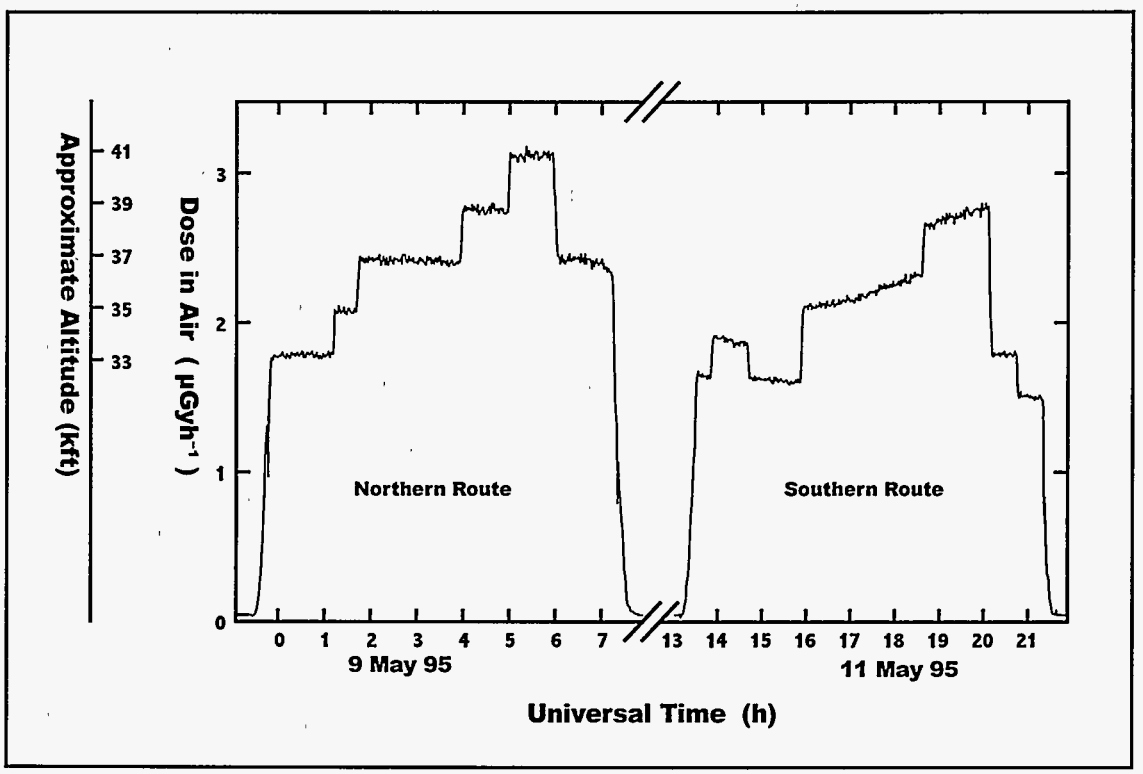

Figure 2.5 Dose rate in air measured by the pressurized argon ionization chamber aboard a Canadian Forces Boeing 707 aircraft as a function of time in hours for the flight from Trenton, Ontario, to Cologne, Germany, on 5/9/95 (northern route) and the return flight on 5/11/95 (southern route).

2.7 ATMOSPHERIC IONIZING RADIATION MEASUREMENTS AT HIGH ALTITUDE USING NASA ER-2 AIRCRAFT

Paul Goldhagen and William Van Steveninck

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 such aircraft would be exposed to roughly twice the levels of atmospheric ionizing radiation as present-day air crews, whose dose equivalents are not yet known precisely.

In collaboration with the NASA Langley Research Center (LaRC), in 1994 we examined the radiation safety aspects of the HSCT (Wilson, et al., 1995). Among our conclusions was that 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. At NASA's request, the National Council on Radiation Protection and Measurements (NCRP) studied radiation exposure and high-altitude flight and in July 1995 published a Commentary on the subject (NCRP, 1995). Among their eight recommendations were:

(1) Average absorbed dose rates and their uncertainty in the altitude range of 9,000 to 24,000 m (30,000 to 80,000 ft) require greater specification.

(2) Additional measurements utilizing currently flying high-altitude aircraft should be made with adequate instrumentation to assist in completion of recommendation (1), (See Appendix A, NCRP Commentary, No.12 for details).

In response to these studies and recommendations, LaRC and EML have developed the atmospheric ionizing radiation (AIR) project. While LaRC develops its AIR environment model computer code, EML will be the principal investigating laboratory for a series of AIR measurements using the NASA ER-2 aircraft as a platform. The primary instruments will be the EML instruments used on the Canadian flights (see Summary No. 2.6), modified to operate at the low air pressure in the ER-2 instrument bays. Results of the measurements will be used as benchmarks to test and validate the AIR environment model code, which can then be used to calculate the dose-equivalent rate at any altitude anywhere on earth at any time in the solar cycle.

It is important to begin the measurements as soon as possible because the solar activity cycle is now at its minimum (resulting in maximum radiation levels) and because decisions concerning the HSCT need to be made within a few years. To make the most complete set of measurements possible with the short development time available, collaborations have been initiated with a number of other radiation measurement groups that have appropriate instruments or are already developing them. Figure 2.6 shows a drawing of the ER-2 and lists the experiments now planned and their locations on the aircraft. Bonner spheres are another name for the detectors of the MNS. The Johnson Space Center (JSC) has a particle telescope that has already been used in space. RMC will fly superheated drop/bubble neutron detectors similar to those it used on the Canadian Forces Boeing

707, and Apřel Enterprises will provide its new REMbrandt™ bubble-detector remmeter. The Boeing Company will provide a small personal dosimeter and a tissue-equivalent proportional counter (TEPC) similar to the one used by DREO on the 707. DREO's TEPC will also be flown. The two TEPCs will be filled at different pressures to provide different microdosimetric site sizes. The German Air and Space Research Institute (DLR) and Kiel University will send a small particle telescope called DOSTEL and plastic nuclear track detectors (PNTDs). The University of San Francisco and the National Radiation Protection Board (NRPB) of Britain will send different types of PNTDs. This suite of instruments exceeds the recommendations in the Appendix of the NCRP Commentary, and fills all the available space in the ER-2. The first flight is scheduled for Fall, 1996.

References

National Council on Radiation Protection and Measurements
"Radiation Exposure and High-Altitude Flight"
Commentary No. 12, NCRP Publications, Bethesda, MD, (1995)

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"
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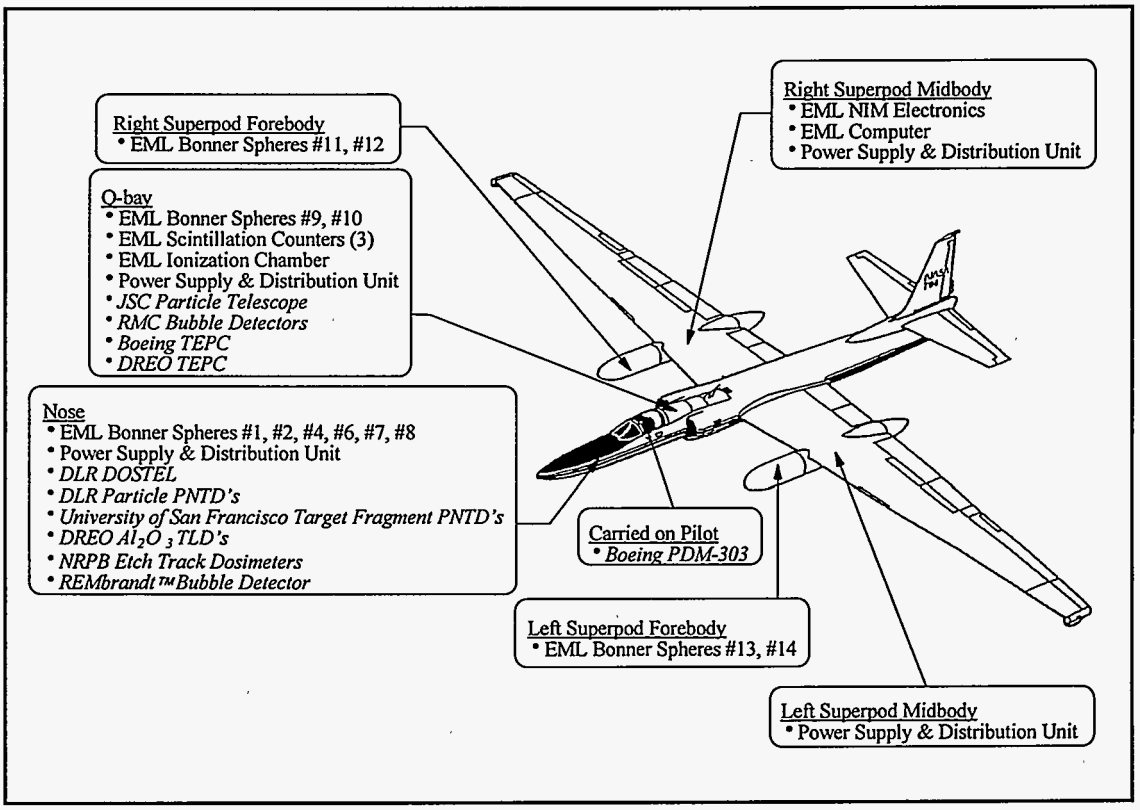

Figure 2.6 Experiment equipment locations on the ER-2 for the high-altitude atmospheric ionizing radiation measurements.

2.8 THE NEUTRON RESPONSE OF Al_2O_3:C , ^{7}LiF:Mg,Cu,P, and ^{7}LiF:Mg,Ti TLDs

Nestor Azziz and Gladys A. Klemic

An investigation of the relative neutron:gamma response of some newly developed thermoluminescent dosimeters (TLDs) was completed in 1995 and presented at the 11th International Conference on Solid State Dosimetry in Budapest, Hungary. The recently developed TLDs Al_2O_3:C and ^{7}LiF:Mg,Cu,P have very high photon sensitivity and are, therefore, promising for use in personnel and environmental dosimetry. For potential application in mixed neutron-gamma fields, we investigated the relative neutron:gamma sensitivity ("k_u") of these TLDs compared to that of the widely used ^{7}LiF:Mg,Ti (TLD-700). Narrow-spectra 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.

The neutron and gamma doses were measured independently using a tissue-equivalent ionization chamber in combination with a compensated Geiger-Mueller detector. 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. TLDs were read out using a manually operated, linearly heated planchet reader. Figure 2.7 illustrates the results. For neutron energies from 0.3 to 6.0 MeV, the k_u of Al_2O_3:C was about one tenth that of TLD-700. The k_u of ^{7}LiF:Mg,Cu,P was about one third that of TLD-700 for the same neutron energy range. All phosphors showed greater response to 14 MeV neutrons, but the relative neutron:gamma sensitivity of the newer phosphors was one third to one fifth that of TLD-700. The results are to be published in Radiation Protection Dosimetry (Klemic, in press).

Reference

Klemic, G. A., N. Azziz, and S.A. Marino
"The Neutron Response of Al_2O_3:C , ^{7}LiF:Mg,Cu,P, and ^{7}LiF:Mg,Ti TLDs"
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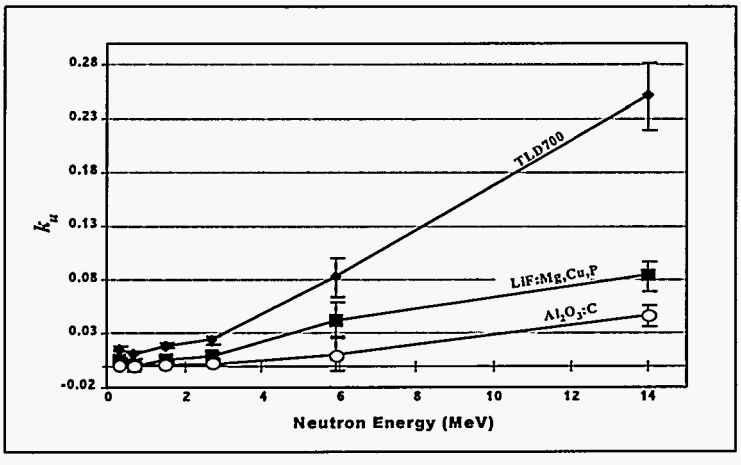

Figure 2.7 Relative neutron sensitivity, k_u (neutron:gamma).

3 ENVIRONMENTAL RADON, THORON AND RELATED AEROSOLS

3.1 OVERVIEW

Alfred J. Cavallo

The 1995 EML Radon Program was active in the following areas:

Quality Assurance Activities

EML, with its extremely stable, NIST-traceable calibrated pulse ionization chambers and its expertise in aerosols and radon progeny measurements, has long been a reference laboratory for radon and thoron measurements. It is a regional intercomparison center for radon, thoron and progeny measurements in the International Radon Metrology Program (IRMP). As a participant in this program, it is one of three regional reference laboratories in the International Intercalibration and Intercomparison Program for Radon, Thoron and Daughter Measuring Equipment (IIIP) of the International Atomic Energy Agency (IAEA). The other two are the NRPB (National Radiation Protection Board), Chilton, UK, and the ARL (Australian Radiation Laboratory), Melbourne, Australia. It has also served as the Quality Control/Quality Assurance Center for the DOE OHER (Office of Health and Environmental Research) Radon Research Program. This work is described in Summary Nos. 3.2 - 3.6.

Experimental Programs

EML's experimental investigations for 1995 occurred in three project areas as follows:

(1) Exposure/Dose Assessment Studies - The risk coefficient for lung cancer due to exposure to radon progeny is based on epidemiology studies of several cohorts of uranium miners. The computed dose to these miners is based on an assumed activity weighted particle size distribution that may neglect important components. Using modern equipment, the atmosphere of a recently opened uranium mine (the Eagle Point Mine at Rabbit Lake in northern Saskatchewan, Canada) has been characterized to determine if the current assumptions on the particle size distribution are valid. A new and potentially important component of the aerosol in uranium mine atmospheres has been discovered. This project is described in Summary Nos. 3.7 and 3.8.

(2) Soil Gas Measurements - This project addressed the following issues: the importance of geologic settings on soil gas radon variations, the relative importance of variables such as permeability and soil temperatures in soil gas transport, and the investigation of the use of the ratio of thoron to radon in soil gas to distinguish between diffusion and advection transport. This investigation is described in Summary No. 3.9.

(3) Detector/New Techniques Development - Measurement technology for this program currently focuses on the development of small and portable devices for indoor radon, and thoron

concentration measurements, and on methods for measuring very low levels of radon gas. This work is described in Summary No. 3.10.

Quality Assurance Activities

3.2 NATIONAL RADON GAS INTERCOMPARISON

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

EML is the USDOE quality assurance center for radon gas and progeny measurements. A program of radon gas intercomparisons began in 1981. The Office of Energy Research, Office of Health and Environmental Research (OHER), funded major programs focused on the health effects of indoor radon. OHER recognized the need for a quality control program to establish the validity of the data obtained through its programs. Thus, OHER mandated the participation of its radon contractors in appropriate EML radon and radon progeny intercomparisons.

The 27th EML Radon Gas Intercomparison was held in April, 1995. The participating groups included three U. S. federal laboratories, one national laboratory, three state laboratories, six universities and ten commercial vendors. There were six foreign participants with three Canadian institutions and one each from Argentina, the Czech Republic and Switzerland.

For the first time, the data summary was decoded to identify the participants and to make this quality assurance information readily available to interested parties. Of the 30 participants, 3 groups were outside $\pm$10% of the EML pulse ionization chamber value of 1040 $\pm$ 20 Bq ^{222}Rn m^{-3}.

An EML report documenting the results of the 1994 radon gas intercomparisons has been issued. (Fisenne, 1995).

Reference

Fisenne, I.M., A.C. George, P.M. Perry and H.W. Keller
"The April 1994 and October 1994 Radon Intercomparisons at EML"
USDOE Report EML-568, October (1995)

3.3 RADON AND RADON PROGENY MEASUREMENT WORKSHOP

Andreas C. George and Keng Wu Tu

A workshop was held at EML November 15-22, 1995 to discuss, intercompare and evaluate radon and radon progeny measurements made in Japan for the assessment of the radiation exposure of the general public. There were ten participants from Japan representing the following laboratories and universities.

- Advanced Research Center for Science and Engineering
- National Institute of Radiological Sciences
- Power Reactor and Nuclear Fuel Development Corporation
- Institute of Environmental Sciences
- Japan Chemical Analysis Center
- Science University of Tokyo
- Research Reactor Institute of Kyoto University
- School of Engineering, Nagoya University
- Gifu College of Medical Technology

Passive integrating monitors consisting of activated carbon collectors, solid state electrostatic collector monitors and nuclear track monitors were exposed in the EML radon test facility under different conditions of radon concentration. Current ionization chambers were tested by three groups representing primary radon test laboratories in Japan. Scintillation cell monitors were tested and evaluated by two participants. Several continuous potential alpha energy concentration (PAEC) monitors were tested under different conditions of radon progeny, airborne particle concentrations and particle sizes. The concentrations of individual radon progeny and PAEC were measured by two participants using alpha spectrometry to count the radioactivity collected for five minutes on membrane filters. At first, both alpha spectrometric measurements deviated from the actual values due to counting efficiency differences and undetermined overlapping of ^{214}Po into the ^{218}Po energy region. The results of the continuous PAEC monitors indicate radon progeny losses by plateout when the particle concentration during testing was less than 5,000 particles cm^{-3}.

The most significant accomplishment during this intercomparison was the intercalibration of the ionization chambers currently used in Japan for their primary radon measurement methodology. The workshop was very productive in terms of assuring standardization of the instruments and methods for measuring radon and radon progeny on an international level.

3.4 INTERCOMPARISON OF INSTRUMENTS AND METHODS FOR MEASURING RADON AND RADON PROGENY

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

An intercomparison exercise for radon and radon progeny instruments and methods was held at EML from April 28 to May 10, 1995. The intercomparison was conducted in the new 30 m^3 radon test facility in accordance with OHER requirements and as part of the International Radon Metrology Program (IRMP). EML, as a reference laboratory for North America and Canada, conducts this intercomparison annually.

Instruments that measure radon with open face and diffusion barrier activated carbon collectors were sent by 13 participants, while 10 sent nuclear alpha track detectors, 9 with short-term and long-term electret/ionization chambers, and 13 sent active and passive commercial electronic continuous monitors. For radon progeny, there were four participants that came in person to take part in the grab sampling methodology for measuring individual radon progeny and the potential alpha energy concentration (PAEC). There were also ten participants with continuous and integrating commercial electronic instruments for PAEC. The results indicate that all the tested instruments that measure radon did well. All instruments and methods for measuring radon progeny by grab sampling techniques performed well. However, when the particle concentration is less than 5,000 cm^{-1}, most of the continuous commercial instruments for measuring PAEC or working level (WL) appear to underestimate the potential risk from radon progeny.

The IRMP provides technical training and advice on the use of state-of-the-art instrumentation for radon, thoron and progeny surveys and for scientific research. The IRMP is involved in the IAEA/CEC Coordinated Research Program on Radon in the Human Environment which involves more than 60 nations worldwide. The main purpose of the IRMP intercomparisons is to ensure consistency in the radon measurements on a global scale.

In the last two years, EML has been involved in preparing the main portion of a document on the design criteria and operational characteristics of radon and radon progeny calibration facilities for the IRMP. Information was collected on radon test facilities operating worldwide. The purpose of this effort was to document the characteristics of different radon test facilities so that the member nations could have easy access for calibration services and for their research needs. An inventory of more than 40 radon test facilities representing primary, secondary and field centers has been compiled and submitted to the IAEA for inclusion in the final document.

Reference

George, A.C., E.O.Knutson, K.W. Tu and I. M. Fisenne
"Intercomparison of Active, Passive and Continuous Instruments for Radon and Radon Progeny"
USDOE Report EML-577, in press

3.5 COORDINATED RESEARCH AND CONSULTATIONS IN INSTRUMENT DEVELOPMENT EVALUATION

Andreas C. George

Instrument development for the assessment of the health risk from radon, thoron and their progeny continues in commercial and monitoring organizations. EML provides its test facilities for the evaluation, upgrading and refinement of instruments to insure good measurement quality. QA radon exposures were conducted with new scintillation cells for the U.S. EPA Risk Management Research Laboratory, the NY State Department of Environmental Conservation, the Pennsylvania Department of Environmental Resources and the NJ Department of Environmental Protection.

Prototype liquid scintillation type activated carbon collectors for radon were calibrated and evaluated for the Radon Testing Corporation of America and Teledyne Isotopes, Inc. Nuclear alpha track detectors were evaluated for several Japanese research laboratories. QA work was conducted to evaluate the primary methods for radon measurements used in Japan. Also, pulse ionization chambers were tested for the Oak Ridge National Laboratory (ORNL) Health Sciences Division. EML provided its radon facilities to the International Academy of Hi-Tech Services for tests on radon progeny dynamics such as attachment, surface deposition and particle size.

As a member of the DOE Radiological Control Coordinating Committee's (RCCC) Radon Subcommittee, Andreas George contributed to the final report on the "Occupational Exposure to Radon, Thoron and their Progeny." Such occupational exposure involves about 3,000 workers. The final document provides an overview of national and international guidance and regulations for occupational exposure to radon and thoron. This Subcommittee makes recommendations for the proper use of instruments and offers a comprehensive program at DOE sites in the assessment, control and recording of occupational exposure.

Andreas George also gave the "Technical Keynote Address" to the American Association of Radon Scientists and Technologists at the 1995 International Radon Symposium in Nashville, Tennessee, on the intercomparison and performance of different instruments for the measurement of radon, thoron and their progeny.

At the request of the U.S. Department of Housing and Urban Development (HUD), Andreas George reviewed the report, "Radon Testing Protocols and Measurement Devices for HUD Assisted Housing." In addition, Mr. George served as a consultant to the American Water Works Association on the impact of water-borne radon to human health during showering.

3.6 INTERNATIONAL INTERCOMPARISON MEASUREMENTS OF RADON PROGENY PARTICLE SIZE DISTRIBUTION

Keng-Wu Tu, Earl. O. Knutson and Alfred J. Cavallo

EML hosted an international intercomparison of measurements of radon progeny particle size distribution during the week of June 12-15, 1995. It is important that progeny particle size distribution be accurately measured because the dose to various portions of the lung is dependent on the size distribution of the particle inhaled. The purpose was to reaffirm quality control procedures and to improve communications among the different groups making the measurements. Participants included three foreign and three domestic laboratories: AEA Technology, the United Kingdom; Isotope Laboratory (IL), University of Gottingen, Germany; Australian Radiation Laboratory, Australia; U.S. Bureau of Mines, Denver Research Center; Clarkson University, Potsdam, NY; and EML, the host laboratory. The participants from AEA Technology, the Australian Radiation Laboratory, and the U.S. Bureau of Mines used the screen diffusion battery technique for this exercise, while the others chose the low pressure impaction methods for this test.

Twenty measurements were made at seven controlled particle size distributions at given radon concentrations. All the participants took samples nearly simultaneously for 5 to 15 minutes. All the samples were alpha counted except Isotope Laboratory's which were determined with a NaI detector for the gamma radioactivity from ^{214}Pb.

At present, data has been received from three groups: AEA Technology used six screen diffusion batteries in parallel and sampled at 6.5 lpm; Isotope Laboratory used a low-pressure cascade impactor (Berner, Model LP130), which consists of eight stages and a backup filter, sampling at 30 lpm; and EML used a micro-orifice uniform deposit impactor (MOUDI, MSP Corporation, MN) which also consists of eight stages and a backup filter, sampling at 30 lpm. A graded screen array (GSA) was also used by EML to monitor the ultra-fine particles in the aerosol.

Because the Isotope Laboratory group determined activity size distributions based on gamma radioactivity of ^{214}Pb only, we chose this progeny size for the comparison. Table 3.1 lists typical results. Most of the data obtained from the two impactors agreed quite well. The diffusion battery system provided broader size spectra for the particles with diameters greater than 100 nm. For the bimodal size distribution particles, only the MOUDI observed minor modes with diameters less than 150 nm.

TABLE 3.1

Comparison of ^{214}Pb Particle Size Distributions from the EML MOUDI, IL BERNER Imactor and AEA Diffusion Battery System.

				EML		Isotope Lab		AEA	
Run #	Rn Conc Bq m^{-3}	CNC++ 1000 cm^{-3}	Dc+ nm	AGMD* nm	GSD**	AGMD* nm	GSD**	AGMD* nm	GSD**
121447	2230	7	80	56	1.58	93	2.86	69	1.98
121600	1991	7		67	2.67	163	2.62	--	--
121700	1784	8		79	1.7	85	3.13	--	--
130900	1894	8	90	--	--	86	1.68	281	3.7
131000	1868	14		84	1.61	89	1.76	119	2.67
131110	1600	16		84	1.6	92	1.64	635	7.58
131405	2156	17	165	174	1.37	172	1.54	446	3.43
131505	1631	18		158	1.56	172	1.54	485	1.94
131610	1233	20		167	1.6	177	1.5	504	1.46
140930	870	6.7	395	374	1.31	356	1.89	504	1.27
141040	939	7.7		375	1.3	380	1.63	504	1.27
141200	776	8.4		393	1.34	395	1.90	494	1.59
141400	896	16	160 & 365 Bimodal	103 375	1.33 1.32	--	--	--	--
						291	2.17	480	1.87
141520	871	14	160 & 365 Bimodal	104 374	1.36 1.34	--	--	--	--
						344	1.63	451	2.23
141620	902	15	160 & 365 Bimodal	83 374	1.78 1.35	--	--	--	--
						289	1.88	497	1.51
150900	2009	10	70 & 400 Bimodal	95 377	1.38 1.32	--	--	--	--
						323	1.81	440	2.81
151000	1694	10	70 & 400 Bimodal	110 386	1.33 1.37	--	--	--	--
						421	1.85	502	1.45
151110	1290	10	70 & 400 Bimodal	100 375	1.34 1.32	--	--	--	--
						383	1.74	505	1.38
151330	1876	11	1200	1277	1.32	1327	1.18	--	--
151420	2050	10		1187	1.41	1373	1.18	--	--

* AGMD: Activity-weighted geometric mean diameter
** GSD: Geometric standard deviation
\+ Dc: Controlled particle modal diameter
++ CNC: Particle number concentration with TSI 3025 condensation nuclei counter

Experimental Programs

3.7 EXPOSURE/DOSE ASSESSMENT

Alfred J. Cavallo, Adam R. Hutter and Keng-Wu Tu

It is critical that the calculation of the human risk coefficient for radon exposure be based on as sound a scientific basis as possible, given the limitations that are inherent in its derivation. This risk coefficient is used to set limits both on occupational and domestic radon levels. Since the uranium mining and processing industry is now quite small, the impact that domestic exposure limits has is actually far more important. Domestic radon exposure is caused by radon contaminated soil gas that is drawn into homes due to normal indoor-outdoor pressure differentials. It was recognized only in the early 1980s that this exposure accounts for more than half of the average estimated dose equivalent in the U.S. population (NCRP, 1987). Thus, the estimate of the radon risk coefficient has been examined carefully by the Committee on the Biological Effects of Ionizing Radiation (BEIR IV, 1988). The current guideline of 4 pCi L^{-1} for radon exposure promulgated by the U.S. Environmental Protection Agency is based in large part on the radon risk coefficient given this group.

This risk coefficient is determined from long-term epidemiological studies of several different groups of miners. The dose received by these miners depends not only on the magnitude of their exposure to radon progeny, but also on the size of the particles to which the radon progeny were attached, the so-called activity weighted particle distribution, as well as on breathing patterns; these factors govern the deposition of particles in the tracheobronchial region of the lung (believed to be the portion of the lung most at risk). Figure 3.1 (ICRP, 1994).shows deposition for a breathing rate corresponding to light exercise, the most important miner breathing pattern. Note that the deposition as a function of particle size is virtually flat from about 150 nm to above 20,000 nm; as long as the inhaled particles were in this range, the details of their distribution have no impact on total dose. Figure 3.2 (ICRP, 1994) illustrates deposition as a function of particle size for a breathing rate corresponding to heavy exercise. In this case, both the magnitude and the shape of the deposition changes compared to the lower breathing rate; not only is the deposition higher overall, but also large particles (those with diameters greater than 1000 nm) have a much higher deposition rate.

Current risk coefficient estimates are based on an assumed particle size distribution in the range of 150 to 250 nm (NRC, 1991). However, based on previous work in an experimental uranium mine (Knutson and Tu, 1995), it was felt that this may neglect a significant fraction of the activity attached to particles with a diameter greater than 1000 nm. To resolve this question, a field trip to an active uranium mine was organized. The aim of this trip was to characterize the mine atmosphere as completely as possible, with particular attention to those locations that might duplicate conditions in old mines. The uranium mine selected was the Eagle Point Mine at Rabbit Lake in northern Saskatchewan, Canada, owned and operated by the Cameco Corporation. (We are especially grateful for the complete cooperation and help provided by Cameco employees.)

Equipment used included:

- MOUDI (Micro Orifice Uniform Deposit Impactor)
- Graded Screen Array
- Associated counting equipment (photomultiplier tubes, computers)
- Alphaguard (radon measurements)
- B&K Multigas Spectrometer (CO_2, CO, H_2O)
- Met 1 (Particle size distributions, particle diameter >500 nm)
- Electrical Aerosol Analyzer (particle size distributions)
- CNC (condensation nuclei counter, particle concentration)

Conditions in the mine were particularly challenging, especially for equipment not specifically designed for field use. Temperature in the mine was about 5° C (outside temperatures were -15° C) with a relative humidity of 80 - 100 %. Water dripped from the mine roof and accumulated on the floor, causing electrical short circuits in some of the equipment. The high ventilation rate needed to keep radon levels acceptably low combined with the high humidity and low temperatures to make the working environment physically stressful in spite of many layers of clothing. Some rough idea of the conditions experienced in this mine can be obtained by examining Figure 3.3, which shows an activity-weighted particle size distribution sample being taken using the MOUDI impactor at a site of active drilling.

Radon levels in the mine ranged from 15,000 Bq m^{-3} (400 pCi L^{-1}, normally off limits for miners) at the top of a working stope (a position at which the ore is being extracted) to 567 Bq m^{-3} (15 pCi L^{-1}) at a drilling site. A condensation nuclear counter indicated that particle number concentration was always greater than 70,000 cm^{-3} everywhere in the mine, even in areas with no mining or other activity. Carbon dioxide and carbon monoxide levels were about four times the levels normally found in indoor air; filters in the overnight samplers were black with what appeared to be soot.

A typical activity weighted particle size distribution for the PAEC, shown in Figure 3.4, was centered over the range of 50-100 nm, with virtually no activity outside this range. Virtually all of the activity is due to the short-lived radon progeny ^{218}Po, which is consistent with the high ventilation rate.

At first, we attributed the aerosol characteristics of the mine atmosphere to the underground use of diesel powered ore carriers and jeeps. However, given the high ventilation rates for the mine (1.3 million cubic feet per minute), it seemed completely unreasonable that a small number of diesel engines could possibly create the high particle concentrations and the high CO and CO_2 levels. The particle size distribution, moreover, was not at all typical of what is observed with diesel engines, which is usually centered in the range of 150 to 250 nm (NRC, 1991).

We then realized that the fresh air pumped underground was heated by burning propane in the airstream at a rate of about 25 lpm with the combustion products being carried into the mine with the fresh air. (Outside temperature, as noted above, was significantly colder than in the mine.) Although propane is a clean-burning fuel, a minute amount of fine carbon aerosol is formed during

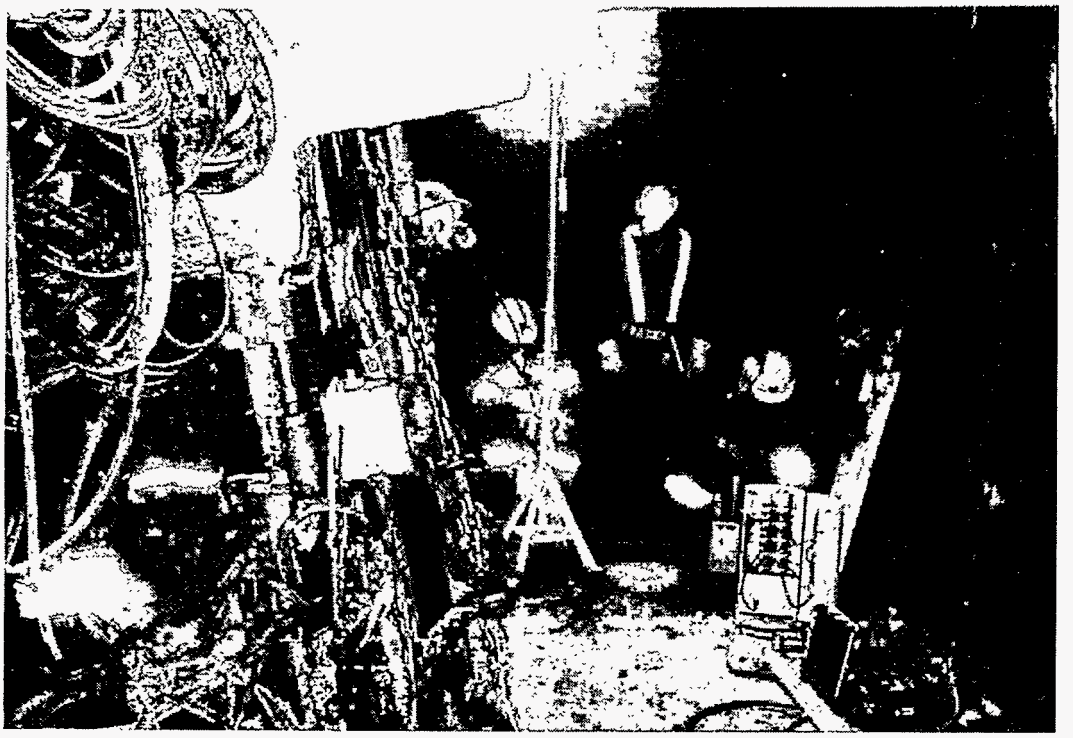

Figure 3.3 Taking a sample with the MOUDI to measure the activity weighted particle size distribution at an active drilling site in the Eagle Point mine 260 meters below the surface.

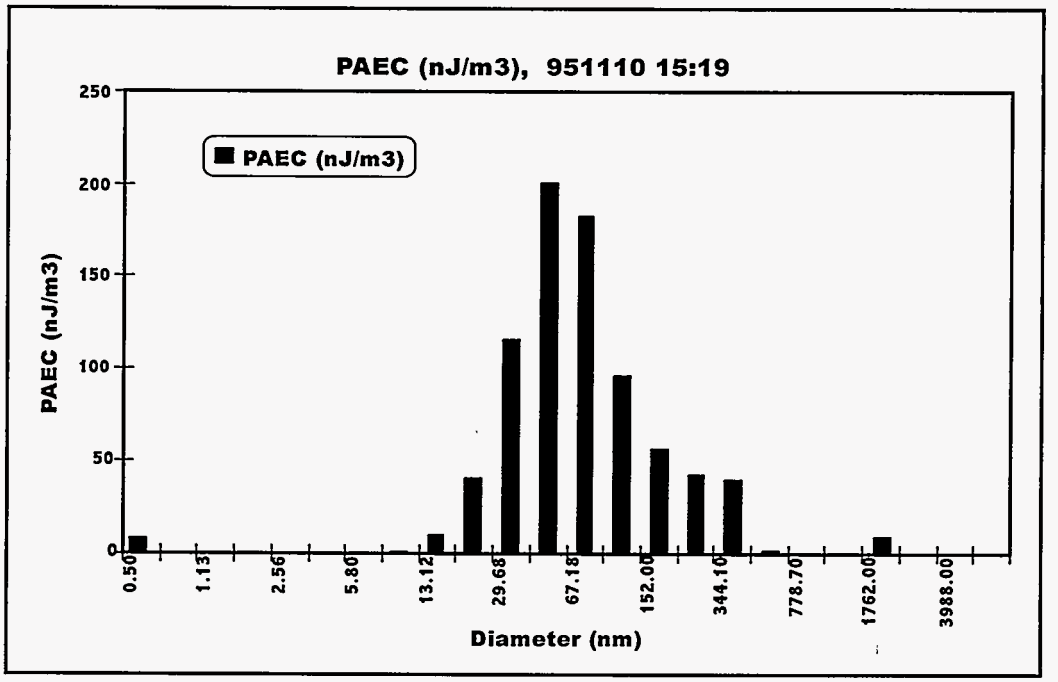

Figure 3.4 PAEC (potential alpha energy concentration, nJ/m-3) as a function of particle diameter(nm). This data was taken on November 10, 1995, at 15:19 in the bolt storage area (located in an alcove off the main decline) about 90 meters below the surface.

3.8 ELECTRON MICROSCOPIC CHARACTERIZATION OF AEROSOLS IN THE RABBIT LAKE URANIUM MINE

Lambros Kromidas, Adam R. Hutter, Alfred J. Cavallo, Keng-Wu Tu, and Peter Roiz

Three aerosol samples analyzed with the electron microscope were collected about twelve inches above the floor of a uranium mine (see Summary No. 3.7) using a radon progeny monitor Pylon AB-5 with an AEP-47 Alpha Detection Assembly. The samples were collected on 10.5 cm^2 polycarbonate Nuclepore filters (0.8 μm pore size). The first sample was retrieved on November 8, 1995 in an isolated location (120 m level) in the mine where very little vehicular activity was taking place. This specimen was accumulated over a period of 24 hours and sampled 1,440 L of air. The second (11/11/95) and third (11/12/95) specimens were collected at the 90 m level, referred to as the bolt storage area. This area was located on the main decline and was characterized by high vehicular activity. The 11/11/95 sample filtered 2,820 L of air over an interval of 47 hours. The 11/12/95 collection sampled 1,740 L of air over an interval of 29 hours.

The filters were sectioned, and a representative portion of each was coated with 10 nm of platinum by a Denton Vacuum Evaporator, DV-502A (Denton Vacuum Inc., Cherry Hill, NJ). Backscatter microscopy of the aerosols was performed by a Robinson Detector (ETPSEMRA, Sydney, Australia) and energy dispersive X-ray microanalysis was performed by the Princeton Gamma-Tech Energy Dispersive Spectrometer, Model 4200, PGT, Princeton, NJ with a two position, light element, lithium drifted silicon x-ray detector.

Results show that the filters contain two major types of particulate aerosols, namely soot and soil. All three samples contained an abundance of soot particles ranging in size from the submicron range to large concatenated clusters many microns in size. Due to their abundance and clustering, obtaining concentration density and size distribution was not possible. X-ray microanalysis of the soot did not reveal any detectable contaminants.

The 11/8/95 sample indicated the concentration of soil particles to be 3,806 particles L^{-1} of air. The average longest dimension of these particles was 1.2 μm. The concentration of soil particles in the 11/12/95 sample was estimated to be 2,870 particles L^{-1} of air. Their average longest dimension was 1.5 μm. These measurements agree with those from the MET-1 particle counter, which indicated average particle concentration of less than 5000 particles per liter for particles with a diameter greater than 0.5 μm everywhere in the mine.

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

Adam R. Hutter

The soil gas Rn project culminated in 1995 with a publication in Health Physics (Hutter, 1995), a paper accepted for publication in Environment International (Hutter, in press), presentations at the Sixth International Symposium on the Natural Radiation Environment (Montreal, Canada, June 1995) and the International Radon Symposium (Nashville, Tennessee, September 27-29, 1995) and organizing and hosting the Sixth International Radon Metrology Program Intercomparison Test and Workshop at EML, June 12-15, 1995. The project addressed issues concerning the importance of geologic settings on soil gas Rn variations; the relative importance of variables affecting soil gas transport; the determination of whether diffusion is the dominant mechanism of soil gas transport, or if other mechanisms may cause unexpected variations in ^{222}Rn concentrations; the investigation of long-term trends and the associated controlling parameters of variations in soil gas ^{220}Rn; and the investigation of the use of ^{220}Rn/^{222}Rn ratio trends to distinguish between steady-state diffusion-dominant migration of soil gas versus advection-dominant.

The project entailed field sampling at three New Jersey sites over a three-year period and resulted in over 1200 soil gas ^{222}Rn measurements, 900 ^{220}Rn soil gas measurements, and 900 soil permeability measurements. The major findings of the project include:

(1) A one-time sample for the determination of soil gas ^{222}Rn and ^{220}Rn concentrations may be accurate only to an order of magnitude due to seasonal effects and the disruption of the soil structure in taking the sample.

(2) Geologic setting strongly influences soil gas Rn dynamics. At a coastal plain site, neither temporal nor spatial variations were observed. At a site near the Reading Prong, strong seasonal and spatial variations in soil gas ^{222}Rn were observed, the timing and magnitude of which defy the widely accepted, yet frequently challenged, conventions concerning soil gas transport.

(3) Diffusive transport cannot explain the timing and magnitude of soil gas ^{222}Rn variations, suggesting that different transport modes may dominate at different times during the year, i.e., diffusion and advection.

(4) The project resulted in the first published long-term trends of soil gas ^{220}Rn. The highest soil gas ^{220}Rn concentrations occur during mid-summer, coinciding with predictions from diffusion-only models. These variations are largely controlled by temperature, both outdoor and soil, with up to 96% of the variability due to temperature alone at some depths (see Figure 3.5).

(5) Permeability measurements, thought to be a major indicator of parameters controlling soil gas Rn variations, show no correlation with ^{222}Rn or ^{220}Rn at any of the sites. This perhaps indicates that soil permeability, as measured, does not accurately reflect parameters thought to control soil gas ^{220}Rn and ^{222}Rn, i.e., soil moisture and porosity.

These results give credence to using radon as a tracer as a means of understanding soil-gas transport in the vadose zone, often important in planning and monitoring remedial actions at many DOE sites.

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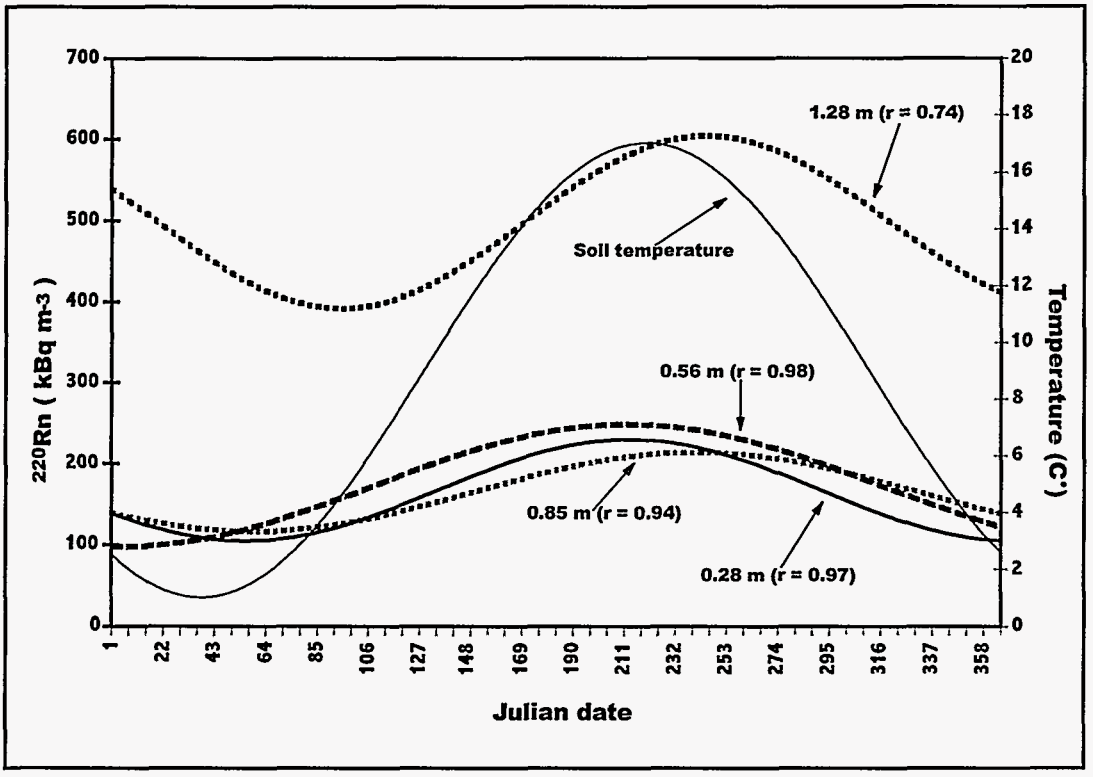

Figure 3.5 Results of soil gas ^{220}Rn profile from three years of data from Chester, NJ site. Soil gas ^{220}Rn and soil temperature lines are data fit following Fourier analyses. Correlation coefficients in legend indicate the r-value between specific soil gas ^{220}Rn depth and the modeled soil temperature.

3.10 SOLID PHASE SCINTILLATION COUNTING OF ^{222}Rn USING SCINTILLATING PLASTIC BEADS

Salvatore C. Scarpitta

Two commercially available plastic beaded scintillation materials are being investigated as alternatives to using 10-15 mL of toluene-based liquid scintillation cocktails for measuring ^{222}Rn by Liquid Scintillation (LS) counting (see Summary No. 6.8). It is well known that by employing LS counting and using low background 20 mL plastic vials, an order of magnitude increase in counting sensitivity can be achieved as compared to gamma counting. For every atom of ^{222}Rn present, three α's and 2 hard β's are emitted at radioactive equilibrium providing a maximum LS count rate of 11 cpm pCi^{-1} of ^{222}Rn (assuming 100% counting efficiency).

Alpha-scintillating plastics, a polyvinyltoluene (PVT) polymer* and polyvinylbenzene (PVB) polymer+, behave similarly to liquid scintillates, producing detectable photons with a wavelength of 400-500 nm due to the interactions of α, β and/or γ emitting progeny, Pb, Bi and Po. These translucent beaded scintillation materials of 100-500 μm diameter, when mixed into small amounts of activated charcoal (or other ^{222}Rn adsorbents), should produce equivalent photon yields compared to a toluene-based liquid scintillant.

Gram amounts of Carboxen, a hydrophobic Beaded Carbon Molecular Sieve (BCMS) material#, with a ^{222}Rn adsorptive capacity that is 2-3 times greater than any activated charcoal, were aliquoted into 20 mL low background plastic scintillation vials and exposed in the EML Radon Chamber for 48 hours at 70% humidity at a radon concentration of 51 pCi L^{-1}. Following exposure, 1 gram of either the PVB or PVT polymer beads was mixed into 1 gram of the BCMS material and counted in a LS analyzer. The results indicate that the PVT material yielded a counting efficiency (CE) of 3 cpm g^{-1} per pCi L^{-1} of ^{222}Rn which was a factor of two greater than the observed CE using the PVB material. The CE obtained after the addition of PVT was itself a factor of 3 greater than the CE obtained when the BCMS/^{222}Rn adsorbent was counted <u>without</u> the scintillating polymer beads. The exposed BCMS material by itself yielded a detectable count rate (37 cpm ± 8%) above background (16 cpm) due to Cerenkov radiation which is produced by β- particle interactions with both the BCMS material and the plastic counting vials. Both polymer beads, by themselves, did not adsorb any ^{222}Rn, as determined from the measured count rates. When 10 mL of Perma-Fluor-V++ (90% toluene, 10% methanol) scintillation cocktail was added to each vial containing the BCMS/^{222}Rn sorbent, the average CE was 15 ± 1 cpm g^{-1} per pCi L^{-1} of ^{222}Rn.

Preliminary results indicate that one gram of beaded PVT polymer, when added to 1 gm of a BCMS sorbent containing adsorbed ^{222}Rn, produces photon signals that can be measured by any LS spectrometer with about one fifth the counting efficiency of a toluene-based liquid scintillant. This reduction is most probably due to photon and alpha attenuation by the BCMS material, which is not translucent. For typical indoor radon concentrations, the combined collection/counting efficiency of 3 cpm g^{-1} per pCi L^{-1} provides activities well above the LLd $_{(1\ hr)}$ of 0.11 pCi (0.24 dpm) for the Packard Counter. The PVT/BCMS mix can be reused by heating at 70° C to desorb both adsorbed ^{222}Rn and water vapor.

* Bicron Corp., 12345 Kinsman Rd, Newberry, OH 44065
+ Duke Scientific Corp. 2463 Faber Pl., PO Box 5005, Palo Alto, CA 94303
♯ Supelco Inc., Supelco Park, Bellefonte, PA 16823
++ Packard Instrument Co., Downers Grove, IL

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4 ATMOSPHERIC AND SURFACE POLLUTANT STUDIES RELATED TO GLOBAL CLIMATE CHANGE

4.1 OVERVIEW

Robert Leifer

Atmospheric and surface pollutant studies related to global climate change at EML primarily focuses on the interaction of aerosols in atmospheric radiative processes. Additional studies investigate transport mechanisms involving natural and anthropogenic tracers. Both anthropogenic and natural atmospheric aerosols 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. For these reasons, investigations pertaining to the role of aerosols in atmospheric processes have become an important area of research at EML.

During the past year, EML's climate research involved the development of an operational aerosol observation system (AOS) for the Atmospheric Radiation Measurements (ARM) Program to help quantify the interaction of aerosols with direct or solar radiation. The AOS consists of a set of aerosol instruments which, when assembled, provide a detailed picture of the surface aerosol structure at the Southern Great Plains (SGP) Cloud and Radiation Testbed (CART) site. The Laboratory provided the engineering design and construction for a special support structure for the AOS. The AOS was installed at the SGP site in October 1995 and is undergoing extensive testing.

Since June 1991, we have continued to provide radon measurements at the Atmospheric/Ocean Chemistry Experiment (AEROCE) site at Tudor Hill, Bermuda, and since December 1990, at the National Oceanic and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory's (CMDL) Mauna Loa Observatory (MLO). Radon observation at the sites provided a unique opportunity for researchers at the University of Maryland to validate 3-d chemical transport model.

EML completed its participation in Project Mohave in which perfluorocarbon tracers were released from potential sources of atmospheric pollutants to investigate the impact of these pollutants on the haze and visibility conditions in the Grand Canyon National Park.

4.2 AEROSOL OBSERVATIONS AT THE SOUTHERN GREAT PLAINS SITE: EQUIPMENT INSTALLATION AND OPERATION

Robert Leifer, Ronald. H. Knuth, Brian Albert, S. Frederic Guggenheim and Hsi-Na Lee

To meet the needs of the ARM program, EML has the responsibility to establish a surface aerosol observation system (AOS) at the SGP site. To minimize the impact of surface agricultural activities at and near the site, sampling of the ambient aerosol occurs at a height of 10 m. To facilitate sampling at 10 m, we have designed a special tilt down sampling stack and manifold for aerosol characterization. The stack is designed for both long-term use and easy maintenance. Figure 4.1a is a photograph of the stack and associated equipment. At the top of the stack is a rain hat containing a stainless steel screen to prevent birds and insects from entering the stack. The tilt down capability allows the screen on the ground to be changed safely and easily. EML provided all the drawings necessary for ARM management to solicit bids for construction of the outside support structure of the AOS. We fabricated the aerosol sampling stack and the stack support structures at EML. The stack support structures were shipped to a subcontractor in Oklahoma for installation at the CART site prior to the arrival of the EML instrument installation team.

The complete system was installed in five electronic racks at EML. This allowed us to custom design many of the specialized components of the AOS at EML and install them in the racks before shipment. After the system was completed and tested, it was completely disassembled and shipped to the SGP for installation in the aerosol trailer. Figure 4.1b is a view of the inside of the trailer showing the sampling inlets and racks.

Six instruments are included in the AOS: a single wavelength nephelometer (λ=530 nm), optical particle counter (OPC), condensation particle counter (CPC), optical absorption monitor (PSAP), a three wavelength nephelometer (λ =450, 550,700 nm) with back scatter capability, and an ozone monitor. The aerosol instruments are connected in a specially designed manifold (Figure 4.2). A 10 μm impactor limits the size of the aerosol reaching the nephelometer, PSAP and CPC. The OPC, which measures the size distribution, is connected through an isokinetic probe to the sampling line before the impactor.

The ozone monitor has its own teflon sampling line and is mounted adjacent to the aerosol sampling stack. As the tubing enters the trailer, a temperature controlled heater gently warms the inlet air to prevent condensation during the air conditioning season. Approximately 1000 L min^{-1} of air enters the stack at a height of 10 m. A heated sampling line inside the stack allows the removal of 150 L min^{-1} of air from the main air stream. This air stream splits into five 30 L min^{-1} streams. Four lines are for isokinetic sampling (particle size preserved in sampling) and one is a non-isokinetic line.

A field data ingestor (Sun Workstation) located in the aerosol trailer interrogates and controls the various instruments and components via RS-232 serial interface ports. In addition, the field data ingestor samples analog signals and performs control functions. We have been collaborating with Richard Eagan of Argonne National Laboratory, who is developing the software for the data acquisition system.

An operator's manual detailing the design and operation of the AOS was written and submitted to the Site Operations Office for approval prior to site personnel taking over the operational maintenance of the AOS.

Trajectory analyses are calculated twice daily for the ARM science team for the characterization of air mass types as they pass over the site. To improve the air mass classification, we are also including observations of the vertical temperature structure and amount of precipitation along the estimated trajectory to evaluate the modification of the upper air mass by local surface pollutants.

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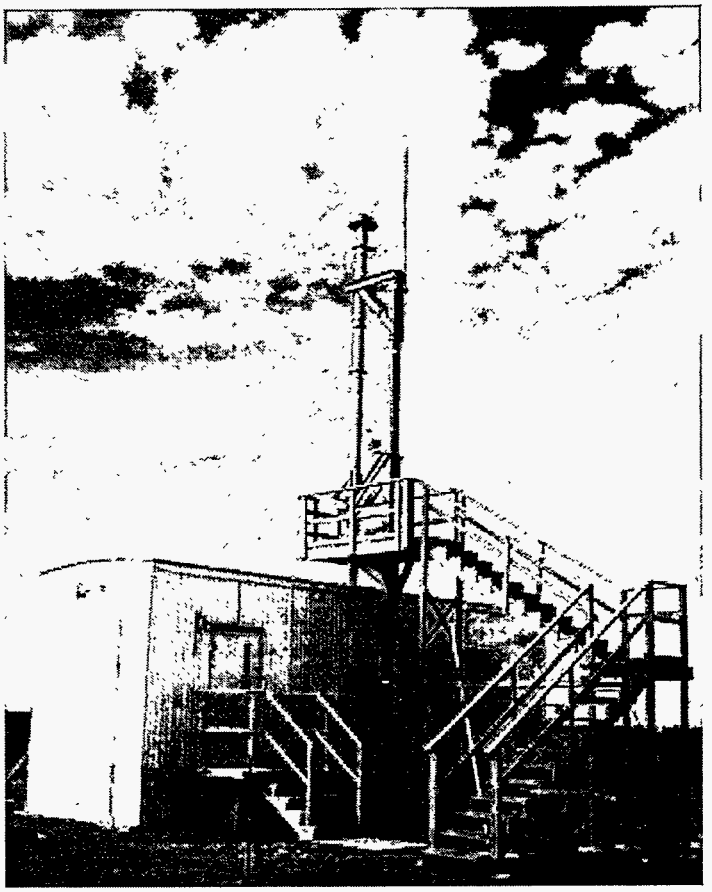

Figure 4.1a Photograph of the aerosol trailer and sampling stack at the Southern Great Plains ARM site.

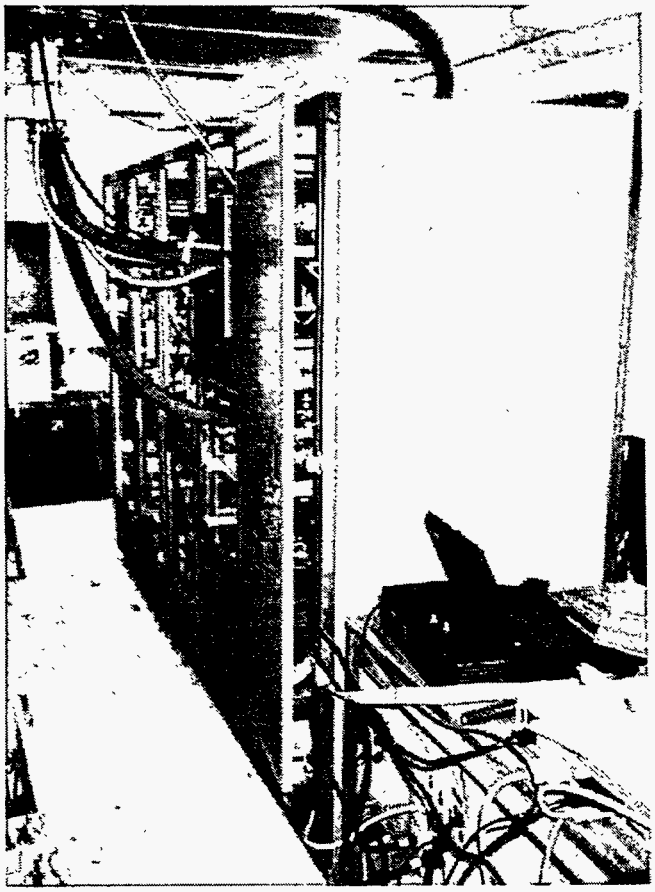

Figure 4.1b Photograph of the inside of the aerosol trailer.

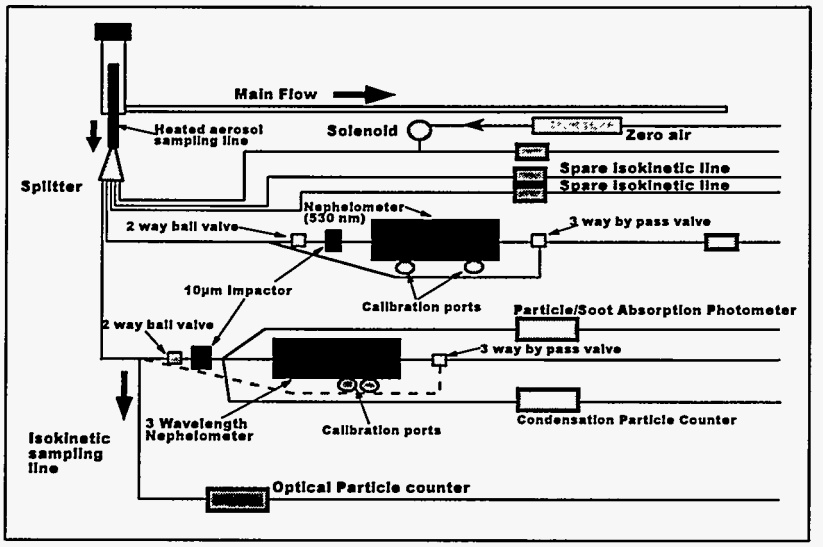

Figure 4.2 A block diagram of the aerosol sampling system (AOS) showing various sampling lines and the major components of the AOS. Air passes through a heated tube controlling the relative humidity of the air stream before splitting into 5 airstreams. Four of the airstreams are isokinetic and one is non-isokinetic.

4.3 ATMOSPHERIC TRACERS

Raymond J. Lagomarsino

Perfluorocarbon tracer concentrations from two-hour surface samples collected in the measurement of haze and visual effects (MOHAVE) experiment (July 1992) at Dolan Springs, AZ, have been transmitted to EPA, Las Vegas, as a database for the development/verification of atmospheric models of the experiment. MOHAVE was designed to determine if pollutants from the Los Angeles basin, El Centro, CA, and a coal-fired power plant located in Lauglin, NV, are transported to Grand Canyon National Park. A different perfluorocarbon tracer was released from each location to mimic the pollutant plume trajectories.

Vertical samples, which were simultaneously obtained with surface samples, were collected at altitudes of up to 500 meters above the surface with a multiport sampler (Polyport) attached to a tether line of a 7.25 m^3 balloon. Only 35% of the anticipated number of above-the-surface samples was obtained because (a) the balloon could not be launched when wind speeds exceeded 10 meters sec^{-1}; and (b) high winds which developed during the flight caused balloon instability. Also, because of weight considerations, no more than two Polyports could be lifted to the desired altitude. Generally, only one sampler was used during each balloon flight of 22 hours. Analytical results performed by EML for the vertical samples have now been completed, compiled and evaluated, and a final report will be sent to EPA, Las Vegas, in 1996.

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4.4 ATMOSPHERIC RADON MEASUREMENTS AT BERMUDA AND MAUNA LOA, HAWAII

Adam R. Hutter and Richard J. Larsen

The EML radon analyzer, installed at the Atmospheric/Ocean Chemistry Experiment (AEROCE) site at Tudor Hill, Bermuda, in June 1991, and at the National Oceanic and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory's (CMDL) Mauna Loa Observatory (MLO) in December 1990 continued to provide high quality useful radon measurements during 1995. A paper was published in 1995 summarizing our radon analyzer and the applicability of the data to air mass provenance determination (Hutter et al., 1995). Additionally, the radon data, when coupled with meteorological data (obtained by Dr. Hal Maring, University of Miami at the AEROCE site, and the CMDL staff at the MLO), provides the scientific community with a unique and comprehensive database which can be used for the validation of atmospheric transport models. For example, the data was recently used by Dr. Dale Allen, University of Maryland, to validate a 3-d chemistry and transport model (Allen, 1996) and by Dr. Natalie Mahowald, National Center for Atmospheric Research (NCAR), to evaluate global chemical transport models (Mahowald, 1995).

The quality of the radon measurements was demonstrated during the first international intercomparison of atmospheric ^{222}Rn analyzers (Colle et al., 1995a; 1995b). The results of the intercomparison indicated the EML analyzer produced radon values that averaged less than 10% different from the NIST standard radon additions (Figure 4.3).

In the future, our radon analyzers will continue to provide a high quality, unique and timely database for validating and evaluating global climate models. Additionally, we are currently evaluating the deployment locations of the radon analyzers.

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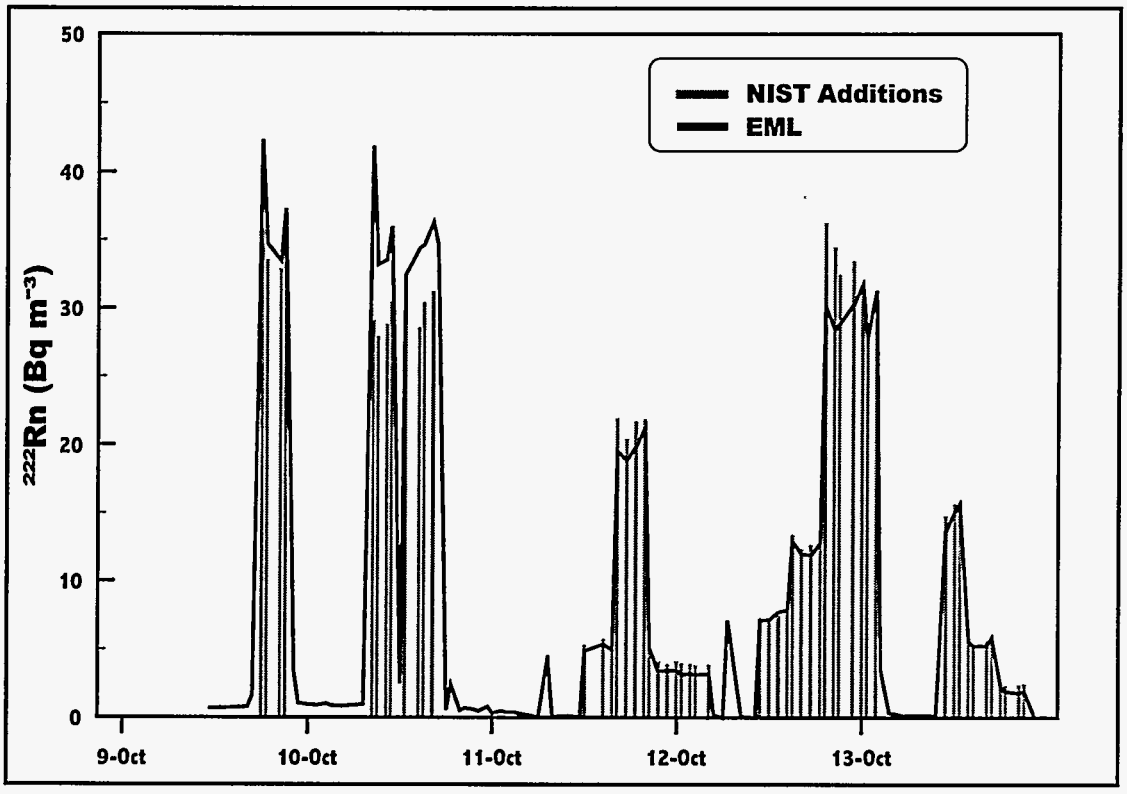

Figure 4.3 The results of the International Intercomparison of marine-atmospheric ^{222}Rn measurements showing EML's results and the NIST standard additions. The arithmetic mean difference for all the measurements between the NIST and EML values is ~ 9%.

5 THE ATMOSPHERIC CHEMISTRY PROGRAM

5.1 OVERVIEW

Richard J. Larsen

The Department of Energy's Atmospheric Chemistry Program (DOE/ACP) was initiated in 1991 to coordinate the atmospheric chemistry research between universities and federal laboratories (OHER, 1993). The program's main focus is on the atmospheric source-receptor sequence from energy-generating sources. The primary objective of the program is to provide DOE with information about the atmosphere that is required for long-range energy planning in fulfillment of the National Energy Policy Act of 1992 (OHER, 1995).

During 1995, the Environmental Measurements Laboratory (EML) participated in ACP by conducting research in the five areas listed below:

1. "EML Sample Archives": The objective of this work is to provide a comprehensive computer generated database that incorporates all of the measurements from relevant EML environmental sampling programs. The database will also include a comprehensive listing of samples that were collected in these programs and are currently archived at EML.

2. "Aerosol Characterization on the Gulfstream Aircraft (G-1)": The objective of this study is to provide aerosol characterization in support of the marine aerosol characterization study (MACS). The goal of MACS is to understand the chlorine budget in the marine environment.

3. "^{222}Rn and ^{222}Rn Progeny Measurements on the Gulfstream Aircraft (G-1)": The objectives of this work are to test and evaluate a newly developed instrument designed to measure charged ^{218}Po in the atmosphere; to obtain high quality atmospheric ^{222}Rn measurements using charcoal trap samples; to provide the ACP community with an additional atmospheric tracer; and to use the ^{222}Rn data to validate a newly developed boundary layer model.

4. "Modeling Trans-Pacific Transport of Combustion Products": The objective of this study is to test the reasonableness of the Global Chemistry Model (GchM), which will be used to assess the impact of Asian combustion products on the United States by simulating ^{222}Rn transport from Asia across the Pacific Ocean.

5. "^{222}Rn Source Terms to the Atmosphere": The objective of this work is to provide a global map of ^{222}Rn source terms to the atmosphere. To generate this map, the parameters that affect the spatial and temporal variations of ^{222}Rn exhalation are studied and differences between ^{222}Rn exhalation measurement techniques are identified.

EML's participation in the 1995 Northeast Field Study represented a significant portion of our field effort in ACP. The home base for the field study was Gabreski Airport located on the eastern end of Long Island in New York State. The study was conducted from August 30 - September 9, 1995 using the DOE Research Aircraft Facility's Grumman Gulfstream-1 aircraft (G-1).

The G-1, shown in Figure 5.1, is a large aircraft equipped with state-of-the-art instrumentation for use in atmospheric studies (Spicer et al.,1994). The overall objectives of the field study were to: evaluate the performance of recently developed instrumentation; quantify the chemical and physical development of the boundary layer between New York City and Boston; conduct a pilot study for urban emissions inventory verification within the Boston area and measure trace gas concentrations over the ocean using a trace atmospheric gas analyzer. Dr. Carl Berkowitz, Pacific Northwest National Laboratory, and Dr. Chet Spicer, Battelle Memorial Institute, were responsible for the overall coordination of the scientific missions. During the field study, 12 flights were conducted, each with specific objectives. There were 6 boundary layer, 2 ocean chlorine, 2 emissions inventory and 2 instrument test flights. EML was responsible for aerosol and ^{222}Rn measurements and collections during these flights. We continue to collaborate with Dr. Berkowitz and Dr. Spicer in the interpretive analysis of the data.

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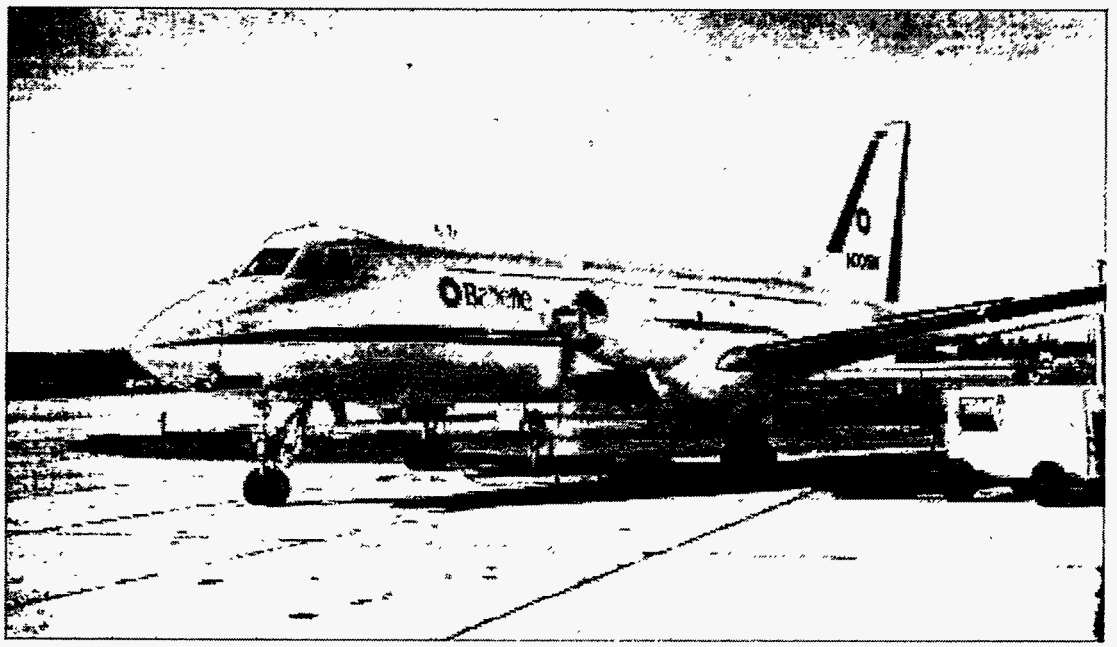

Figure 5.1 The DOE Research Aircraft Facility's Grumman Gulfstream-1 Aircraft (G-1).

5.2 EML SAMPLE ARCHIVES

Robert Leifer and Nita Chan

In recent years there has been a request by the atmospheric modeling community for a complete computer database of all of EML's stratospheric and upper tropospheric radioactivity measurements conducted during the period 1957 through 1983 under the auspices of the U.S. Atomic Energy Commission (AEC), the Energy Research and Development Administration (ERDA) and the Department of Energy (DOE). These data were collected as part of Projects ASHCAN, STARDUST, and AIRSTREAM, collectively known as the High Altitude Sampling Program (HASP), to investigate the impact of the injection of radionuclides and stable compounds into the stratosphere. More than 20,000 filters were obtained during this period and analyzed for up to 20 different radionuclides. In 1995, we completed the first phase of the archive program, which focused on entering all the data from Projects ASHCAN, AIRSTREAM and STARDUST into a radionuclide database (RANDAB) which will be available on-line within the next few months through DOE's Carbon Dioxide Information Analysis Center (CDIAC) at the Oak Ridge National Laboratory. An additional file on stratospheric tritium samples, collected by Dr. Allen Mason of Los Alamos National Laboratory (LANL), during Project AIRSTREAM missions, will be added to RANDAB.

In addition to the radionuclide database, EML's compilation of stratospheric trace gas concentrations has been completed and is available on-line through DOE's CDIAC at:

http://cdiac.esd.ornl.gov

This database, called TRACDAB, contains information on more than 1000 samples, each analyzed for one or more of the following gases: CCl_3F, CCl_2F_2, CCl_4, N_20, SF_6, CO_2, CH_4, CH_3CCl_3, and COS. These data are potentially useful for the development and verification of climate models and understanding stratospheric transport processes. Furthermore, these data may also be useful for modeling the future atmospheric impact of a projected new fleet of stratospheric flying aircraft.

5.3 AEROSOL CHARACTERIZATION ON THE GULFSTREAM AIRCRAFT (G-1)

Robert Leifer, Lambros Kromidas, Brian J. Albert and Richard J. Larsen

To provide aerosol characterization on the G-1 aircraft in support of the marine aerosol characterization study (MACS) on Long Island during August 1995, EML had to design a customized aerosol sampling package. This package contains: (a) a newly designed impactor to characterize the aerosol composition and particle size; (b) a new aircraft aerosol probe; (c) a size distribution instrument; and (d) an integrating nephelometer. A mass flow controller, vacuum system and computer completes the aerosol sampling package. The major objective of the MACS is to understand the chlorine budget in the marine environment. Chlorine is lost from salt particles through reactions with sulfuric or nitric acid. The loss in chlorine has never been fully quantified as a function of aerosol size.

Aerosol Sampling Inlet Design. One of the major concerns when sampling aerosol on a moving platform such as the G-1 is whether the sampled aerosol is representative of the true ambient aerosol. To this end, we have collaborated with Dr. Arnold Muyshondt, University of Arkansas, in the design of a shrouded aerosol probe for the G-1 aircraft (Figure 5.2). To evaluate the performance of this probe, a size distribution instrument located inside the G-1 sampled the aerosol from a manifold that was connected to the inlet of the shrouded probe. A second size distribution instrument mounted directly on the G-1 in the free airstream provided a reference for comparison. The size distribution instrument that sampled from the shrouded probe worked well and provided data with a time and spatial resolution of one minute or 3 km, respectively. The data from the other size distribution instrument will be provided to EML shortly and the performance of the shrouded probe will be determined.

Parallel Impactor (4PI) Study. A new rotating drum impactor, designed at EML, was evaluated on the G-1 during the MACS. This impactor, known as the four-parallel-impactor (4PI), has the capability of collecting four aerosol samples simultaneously. Controlled by a small computer, the impactor can sequentially collect more than 50 samples on a single drum for Scanning Electron Microscopic (SEM) analysis. Two of the drums were selected to have an aerodynamic cut diameter of 0.18 μm and two were chosen to have an aerodynamic cut diameter of 1.0 μm. The sampling time of the impactor can be determined from the counts of the size distribution analyzer for collections at specific times, or the sampling time can be driven by an independent instrument signal such as from a pressure transducer (altitude) or relative humidity sensor, for both in and out of the marine boundary layer sampling. The signal from real time instruments, such as ozone and sulfur dioxide analyzers, can also control the sampling time of the impactor. This impactor uses the same jet design as our single jet impactor. The individual jets of the 4PI are replaceable and allow for the collection of different aerosol sizes. Using our on-board computer, we can adjust the sampling times to collect sufficient giant sea-salt particles for SEM/x-ray analysis.

This impactor weighs about 0.5 kg and is driven by a stepping motor. The aerosols are pulled through a circular jet and impact onto a 5 cm diameter cylinder constructed of aluminum and covered with a coated foil (Apiezone grease or sticky tape). A small d.c. operated vacuum pump maintains a flow of 1.0 L min^{-1} through the impactor. At this flow rate and at STP the impactor has a

calculated 50% cut diameter (the size at which 50% of the particles are collected on the stage) of 0.18 μm using the smaller jet. As the altitude increases, the 50% cut diameter shifts to lower sizes. The collection cylinder from this impactor can be removed easily and placed directly in the SEM chamber, either coated or uncoated, for observation and chemical analysis.

An aerosol sampling system was designed, built and tested during this past year. The equipment was mounted in an aircraft rack used on the G-1. Our shrouded aerosol probe was mounted in one of the aircraft windows adjacent to the aerosol sampling rack. The system consisted of a 4PI, six channel optical particle counter (OPC), integrating nephelometer, computer data logger and controller, vacuum pumps and mass flow controllers. The system was fully automated and pre-programmed for aerosol sampling during each flight. A single switch was turned on at the beginning of every flight.

Figure 5.3 illustrates an example of the real-time aerosol observations obtained during the September 4, 1996 marine flight. The aerosol scattering coefficient (σ_{sp}) and aircraft sample altitude are plotted as a function of time. The lowest σ_{sp} (cleanest air) correlates with the highest altitude. Large variations in the σ_{sp} occurred during the flight, reflecting aerosol inhomogeneity, even at constant altitudes.

More than 200 samples were collected for SEM/x-ray analysis from the 4PI. We used the OPC to determine the sampling time for the 4PI. The 4PI, OPC and nephelometer performed well. Data reduction and analysis were started and should be completed during FY 96. Especially important to this research project is the comparison of the ambient OPC measurements with the sampling manifold OPC. These data will tell how well the shrouded aerosol sampling probe performed.

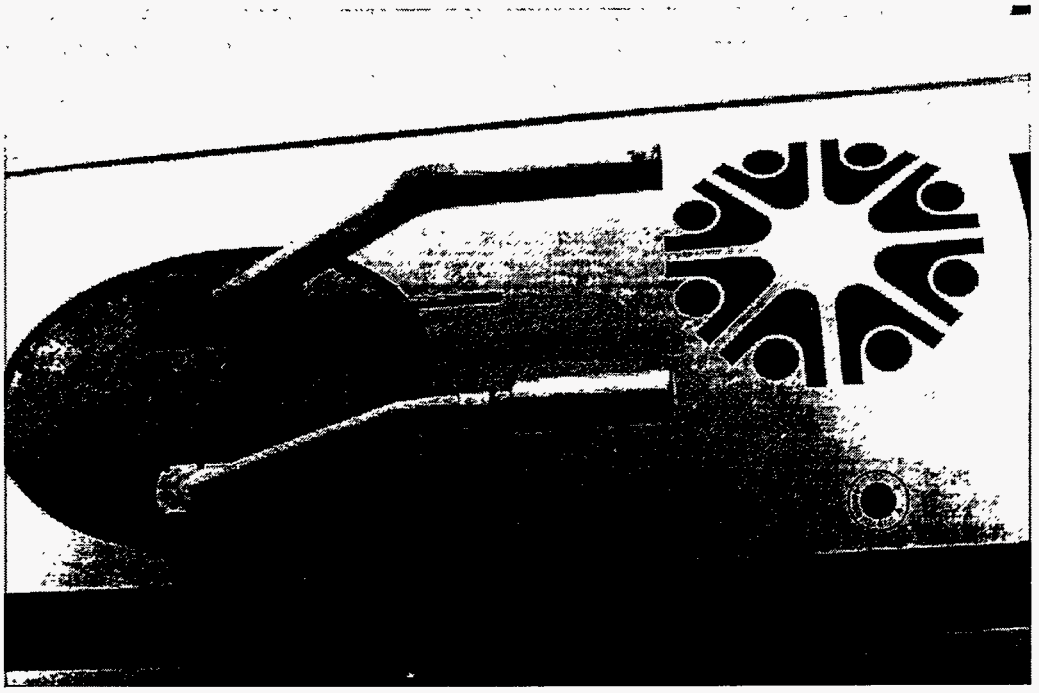

Figure 5.2 View of the G-1 side window containing the radon sampling probe (top probe), the trace atmospheric gas analyzer (TAGA) probe and shrouded aerosol probe (bottom probe).

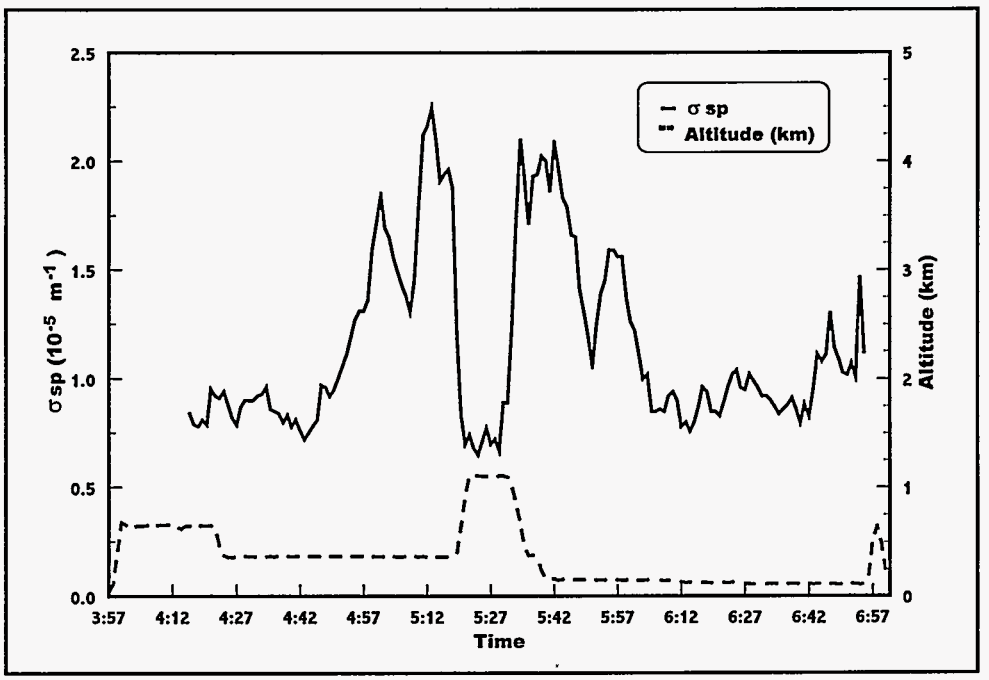

Figure 5.3 A time dependent plot of the aerosol scattering coefficient (σ_{sp}) and G-1 altitude for the September 4, 1995 marine flight.

5.4 ^{222}Rn AND ^{222}Rn PROGENY MEASUREMENTS ON THE GULFSTREAM AIRCRAFT (G-1)

Richard J. Larsen, Brian J. Albert, Harold L. Beck, Norman Y. Chiu, Isabel M. Fisenne, Hsi-Na Lee, Vincent C. Negro, Salvatore C. Scarpitta, William Van Steveninck and Scott B. Wurms

During the 1995 Northeast Field Study, EML successfully used two systems to measure and collect ^{222}Rn in the atmosphere. EML has designed, developed and constructed a new instrument called the "Radgrabber", a compact near real-time radon progeny monitor capable of making precise measurements of charged ^{218}Po from which radon concentrations are inferred. 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. 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 troposphere. Because of neutralization processes, it is believed that the effective half-life of charged ^{218}Po is even less than three minutes, making it more representative of the local radon concentration. To evaluate and calibrate the Radgrabber, charcoal trap samples were collected using a newly constructed semi-automated ^{222}Rn gas sampler.

The specific objectives of EML's ^{222}Rn research were to test and evaluate the Radgrabber; to obtain high quality atmospheric ^{222}Rn measurements using charcoal trap samples; to provide the ACP community with an additional atmospheric tracer; and to use the ^{222}Rn data to validate a newly developed boundary layer model.

Radon-222, a radioactive noble gas with a half-life of 3.8 days, is produced from the decay of ^{226}Ra in the earth's crust. The gas diffuses from the soils of the continents into the atmosphere. Since ^{222}Rn is conservative in the atmosphere and is characteristic of air masses that originate over continental areas, it serves as an excellent tracer to indicate and/or verify the presence of continental air, to estimate the transport time of that air from source to receptor and to validate three-dimensional atmospheric transport models. Because the source term of ^{222}Rn is fairly well known, vertical profile measurements of ^{222}Rn over continental regions are used to determine boundary layer parametizations.

The Radgrabber is a unique instrument using concepts never attempted before. The principle of operation is based on the collection of positively charged ^{218}Po atoms directly on the active surface of a solid state alpha detector by using a strong electrostatic field. Alpha spectroscopy is then used to register the electrically charged ^{218}Po. To collect the charged ^{218}Po requires that the entire front-end electronics be referenced to high voltage, which for the ACP flights was 20 kV. Thus, the alpha detector, amplifiers, pulse height analyzer, and the front-end "TattleTale" computer are all referenced to high voltage. These high voltages permit the unit to sample air up to approximately 30 ft^3/minute, which enhances sensitivity. The air flow is provided using aircraft RAM air through a 2" diameter sampling probe. The flow is controlled on exiting the instrument by a mass flow controller. Power for this section is provided by a battery pack which is also referenced to the high voltage and provides about 75 hours of operation. In addition to this high voltage section, there is also a low voltage section consisting of another computer, several sensors, and output signals. The sensors

measure temperature, humidity, both current and voltage from the high voltage supply, and mass flow, while the signals control a display and the electronics in the high voltage section. To control these sections requires special optical techniques to isolate the high voltage. In addition, both computers share data via this optical link. Since two computers are involved, and one controls the other, the software for the Radgrabber was a major effort. However, the end result is a set of very reliable routines that can automatically correct for problems during the flight. If required, the low voltage computer can stop the high voltage computer, clear its programs, and reload new routines from its memory. In operation, the display gives both 3 and 10 minute running averages of the detected ^{218}Po, while for reliability both computers maintain a data file which is read at the end of the flight.

This compact analyzer was designed for use aboard the DOE/ACP G-1 research aircraft, as well as aboard smaller aircraft where space and payload are limited. The instrument is cylindrical in shape with a length of about 1 m and diameter of about 15 cm. The total weight of the instrument, including the sampling probe and mass flow controller, is ~ 36 lbs (probe ~ 6 lbs, instrument ~ 15 lbs; flow controller ~ 15 lbs). The total power required is minimal, 2.5 amps at 28 V.

The prototype instrument was installed and tested aboard the G-1 (Figure 5.4) during the late August-September 1995 flights based out of Long Island. Although the amplifier is compensated for bright sunlight, one effect that was not anticipated was the chopping of the light by the propellers which creates noise pulses. This problem, for which the term "prop-chop" was coined, was resolved by installing a black baffle. Preliminary data indicate that the Radgrabber is reliable and responsive. Further characterization of the Radgrabber is ongoing at Floyd Bennett Field (see Summary No. 5.5).

An automated ^{222}Rn sampler was designed and constructed at EML to collect up to eight ^{222}Rn charcoal trap samples per flight. The sampler was designed to minimize the effort of manually collecting ^{222}Rn charcoal traps during flight by automating the sampling process. The sampler consists of two sections, one holding eight charcoal traps in a dry ice bath, the other containing rotary and solenoid valves, a mass flow controller, a vacuum pump, and a data acquisition computer. During 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 solenoid valves, vacuum pump, and mass flow controller are operated in the appropriate sampling sequence and the mass flow is measured during a 10 minute sample collection. The sampler is then reset and is ready for the next sampling cycle. After each flight, the manual valves on each trap are closed and the two sections of the sampler are disconnected, allowing for the removal of the individual traps which are sent back to EML for analysis.

During the 1995 Northeast Field Study, ^{222}Rn was collected on 160 g capacity charcoal absorber traps that were initially designed for aircraft sampling in the stratosphere (Harley and Harley, 1965). After each flight, the sampled traps were delivered to EML for analyses. The ^{222}Rn was desorbed into, and measured in, the EML ^{222}Rn pulse ionization chambers, a primary ^{222}Rn measure-ments facility described by Fisenne and Keller (1985). The lower limit of detection at the 95% confidence level for the charcoal trap measurements in the pulse ionization chambers was 0.17 pCi or 6.3 mBq. The traps were returned to the field for use on subsequent flights.

To illustrate the performance of the Radgrabber and the semi-automated ^{222}Rn gas sampler, Figure 5.5 presents the results from the September 3, 1995 flight, a boundary layer mission. Figure 5.5 also depicts the flight path, a box pattern typically used in boundary layer study flights. The circles, numbered sequentially, indicate significant changes in the flight path. Figure 5.6 presents the data obtained during this flight from the Radgrabber and from the analyses of traps collected using the semi-automated ^{222}Rn gas sampler. The figure illustrates the continuous ^{222}Rn record obtained from the Radgrabber and the discrete data obtained from the analyses of the trap samples. As expected, the ^{222}Rn concentration varies inversely with height with a maximum value of 2.3 Bq/m^3 at 348 meters and a minimum concentration of 0.41 Bq/m^3 at 1886 meters. These ^{222}Rn measure-ments are currently being used to determine boundary layer parametizations (see Summary No. 5.6).

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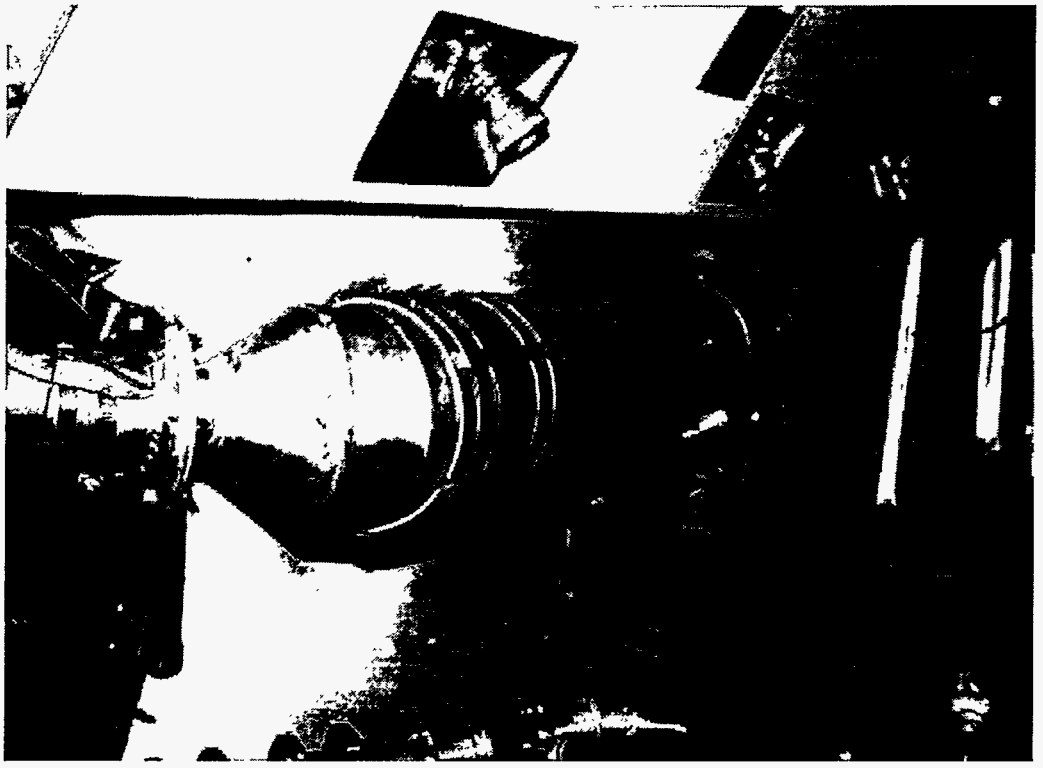

Figure 5.4 The Radgrabber installed on the G-1.

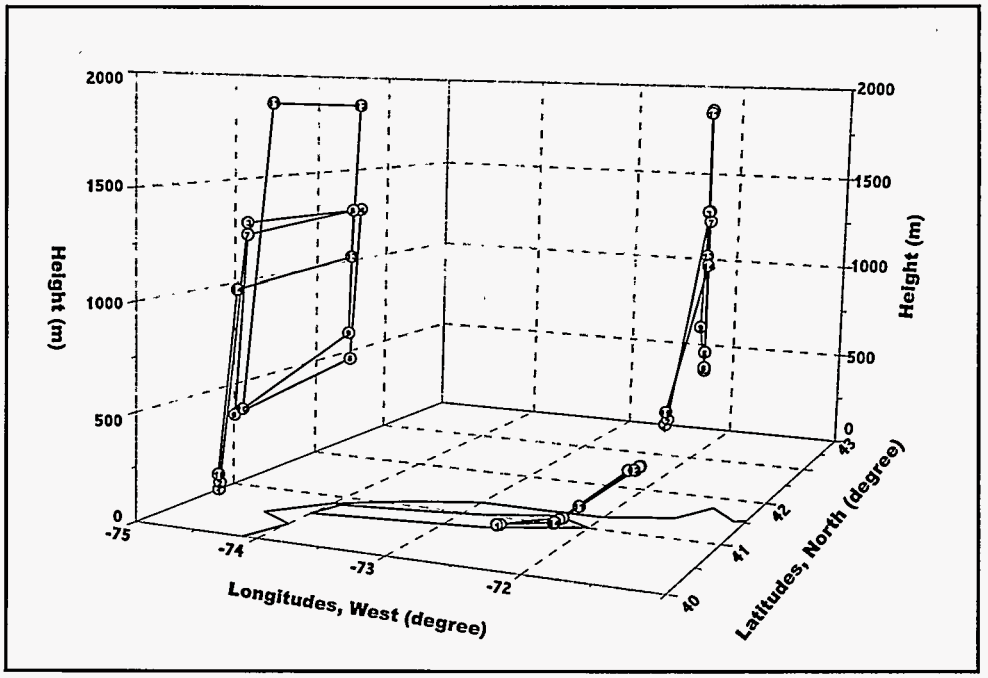

Figure 5.5 The flight path during the September 3rd, 1995 mission.

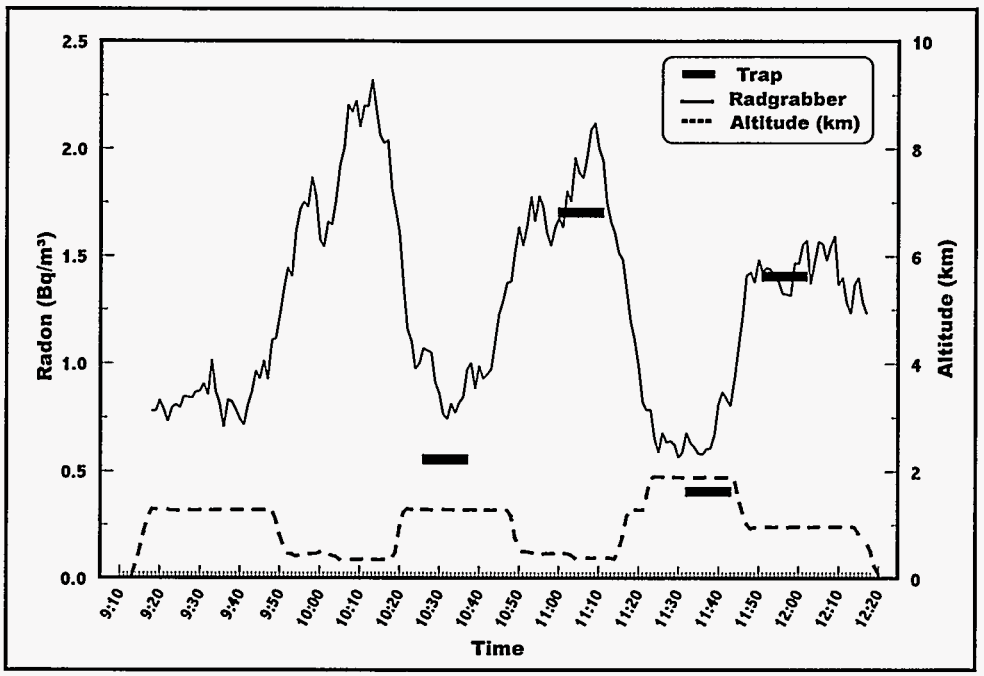

Figure 5.6 Time dependent ^{222}Rn concentrations (Bq/m^3) obtained from the Radgrabber and from trap samples collected during the September 3rd, 1995 mission and the altitude (km) of the G-1.

5.5 CONTINUED TESTING AND EVALUATION OF THE RADGRABBER

Vincent C. Negro, Norman Y. Chiu, Richard J. Larsen, Scott B. Wurms and Cecilia Breheny

The continued testing and evaluation of the "Radgrabber" is required to fully maximize the operational performance of this newly designed instrument (See Summary No. 5.4). Unfortunately, it is difficult to test and evaluate the instrument in the laboratory or inside a test chamber. The ^{222}Rn progeny interact with the many available surfaces, altering the $^{222}Rn/^{218}Po$/charged ^{218}Po relationships. We, therefore, must restrict our testing of the instrument to areas removed from local surfaces. During aircraft flights, the instrument samples air in the free atmosphere, away from local surfaces, but flight time is limited and expensive. An alternate testing platform was set up in the fall of 1995 (Figure 5.7). The Radgrabber was mounted to a 40 ft (~13 m) adjustable ladder which was mounted to the rear of EML's field van. The Radgrabber was hoisted to a height of 37 ft. (~12.3 m) above the ground and was oriented into the wind. In addition to the Radgrabber, a Davis Weather Monitor II wind vane and temperature/dewpoint probe and a sampling inlet for an EML ^{222}Rn gas analyzer, which was located inside the van, were also secured to the top of the ladder. Using this setup, we are able to operate the Radgrabber away from local surfaces and continuously measure the ^{222}Rn concentration and meteorological parameters of the air mass being sampled. The data obtained from the initial testing of this configuration at an open field in Floyd Bennet Field, New York City, is encouraging and continued tests will be conducted during 1996.

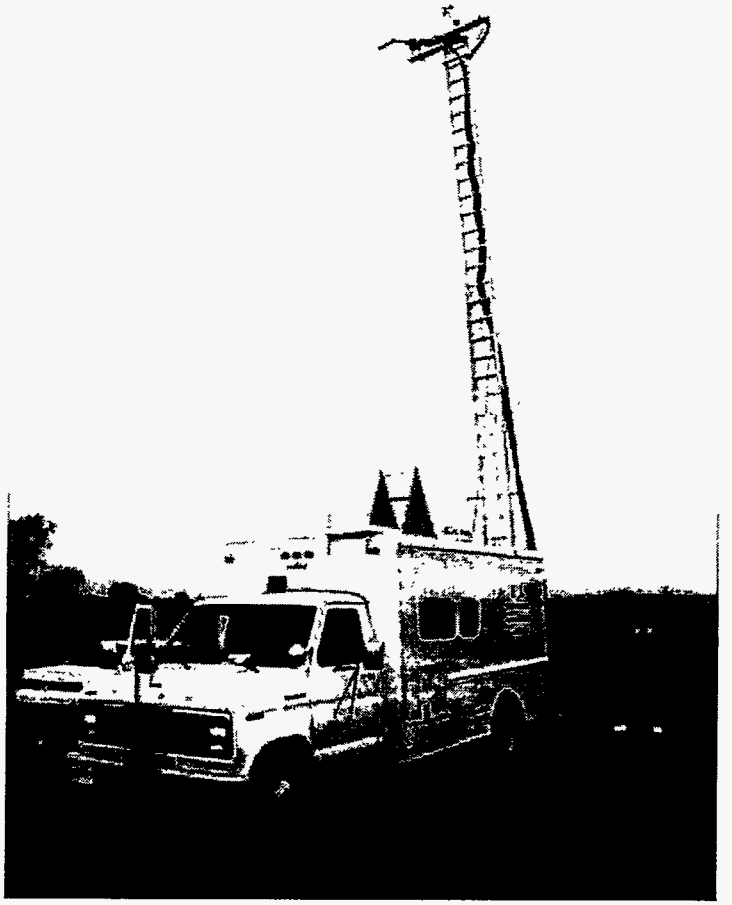

Figure 5.7 Test and evaluation platform for the Radgrabber.

5.6 AN ADVANCED NUMERICAL MODEL FOR CALCULATING VERTICAL PROFILES OF ^{222}Rn AND VERTICAL DIFFUSIVITY IN THE ATMOSPHERIC BOUNDARY LAYER

Hsi-Na Lee

The ability to accurately calculate the vertical profiles of ^{222}Rn and its daughters is very useful for studying the mechanisms of atmospheric transport and for estimating the variations in the background levels of these airborne radionuclides. A time dependent diffusion equation had generally been used to predict the profiles of ^{222}Rn and its daughters at the measurement sites under specified atmospheric conditions. Horizontal advection was neglected under the assumption that the exhalation rates of ^{222}Rn were uniform over large areas. Thus, the vertical profiles of these radionuclides were strongly related to the ^{222}Rn exhalation and atmospheric turbulence which played an important role for vertical diffusion. The vertical diffusion coefficient is quite variable and changes with height, wind shear and atmospheric stability. In order to accurately calculate the vertical profiles of ^{222}Rn and its daughters, the vertical profile of the diffusion coefficient should be precisely determined and the numerical method for solving the time dependent diffusion equation should minimize the numerical errors. To accurately simulate the vertical profiles of ^{222}Rn, an advanced numerical model using the highly accurate spectral numerical technique was developed. Vertical profiles of ^{222}Rn in the atmospheric boundary layer were calculated for several typical profiles of the turbulent diffusion coefficient. The best profile of vertical diffusivity is chosen from the profiles of the diffusion coefficients that produced the best agreement between the calculated profiles of ^{222}Rn and the measured profiles. This modeling technique is being used to study boundary layer parametization using the vertical profiles of ^{222}Rn obtained during the 1995 Northeast Field Study (see Summary No. 5.4).

5.7 MODELING TRANS-PACIFIC TRANSPORT OF COMBUSTION PRODUCTS FROM ASIA BY USING A GLOBAL CHEMISTRY MODEL

Hsi-Na Lee

The long-range transport of Asian combustion products across the Pacific Ocean is of current interest to the scientific community. Asian fossil-fuel consumption and emissions are expected to continue to grow rapidly in the future. These products, when emitted into the atmosphere and transported globally, could have a significant effect on the global climate. By modeling the transport of these pollutants, the current and future trends and the potential impact of these pollutants on the United States can be estimated. A three-dimensional Global Chemistry Model (GChM) which was derived from the original code, PLUVIUS, developed by Dr. Jeremy Hales, ENVAIR, Kennewick, WA, was chosen in order to quantify this impact and to understand the transport processes. The model applies a Eulerian frame and is capable of simulating complex chemistries and other source-receptor behaviors for any specified number of pollutants. In 1995, a joint modeling effort was established between Dr. Lee and Dr. Hales to examine the model's performance for simulating ^{222}Rn transport from Asia across the Pacific Ocean. The transport of ^{222}Rn from Asia should closely mirror the transport of Asian combustion products. Figure 5.8 presents the results of one of the simulations, showing the transport of radon crossing over to the Pacific Ocean and reaching as far as the West Coast of the United States. The collaboration between Dr. Lee and Dr. Hales is expected to continue in the future.

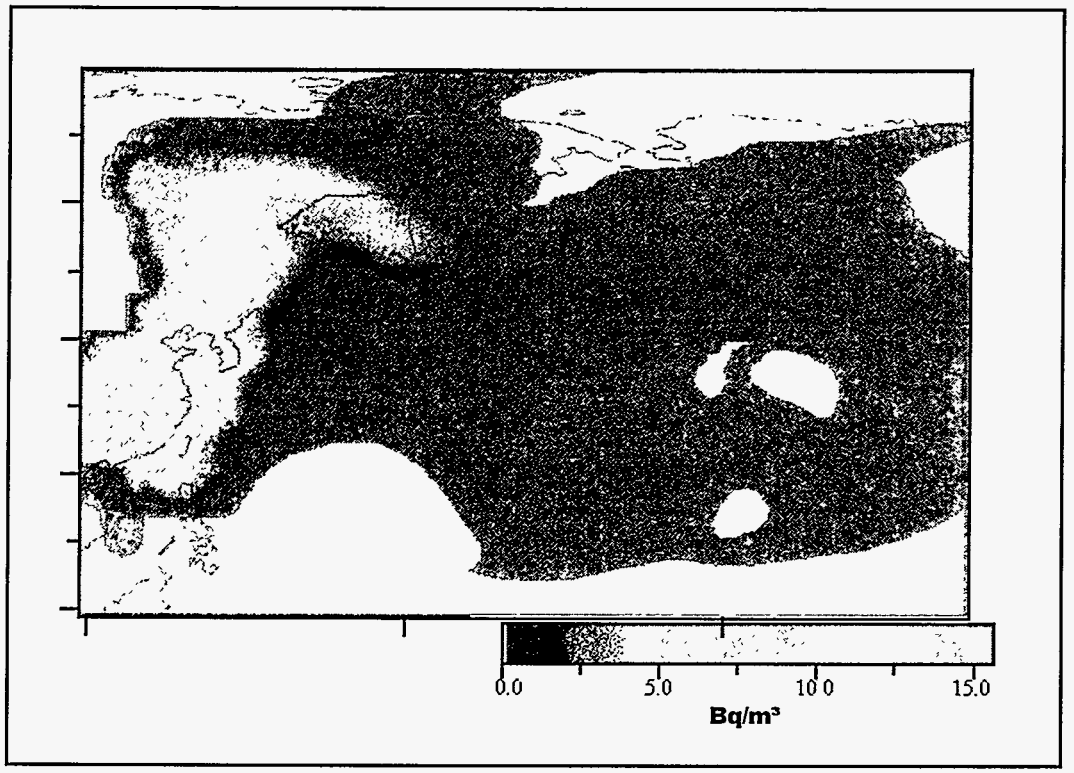

Figure 5.8 A simulated distribution of ^{222}Rn emitted from China after 30-days.

5.8 RADON SOURCE TERMS TO THE ATMOSPHERE

Adam R. Hutter

Atmospheric ^{222}Rn data is used by modelers to evaluate and validate the transport component of General Circulation Models (GCM). Currently, a value of 1 ^{222}Rn atom cm^{-2} sec^{-1} flux rate is used as the source term to the atmosphere for every global land mass. There is a need for better defined and more realistic emission rates of ^{222}Rn, reflecting global differences in, among other parameters, climate and rock and soil type. Working with researchers at the New Mexico Institute of Mining and Technology, modeling ^{222}Rn flux from databases of soil texture, surficial geology and soil moisture was considered to be the only viable approach to solving this problem.

One aspect of developing realistic global estimates of ^{222}Rn emission to the atmosphere involves understanding seasonal variations in ^{222}Rn flux, such that a set of global ^{222}Rn emission rate databases may be needed in order to take into account the effects of global differences in temperature, precipitation and other climate related parameters on the ^{222}Rn flux. Towards this end, a sampling project in a homogeneous setting (New Jersey coastal plain province sands and gravels) was initiated to study seasonal changes in the ^{222}Rn flux to the atmosphere. These findings were presented at the 1995 Spring Meeting of the American Geophysical Union held in Baltimore, MD (Hutter, 1995). Figure 5.9 shows the results of the ^{222}Rn flux measurements and a model based on a data fit following a Fourier analysis. This figure shows the ^{222}Rn flux to be about three times higher in the summer than during the winter at this site, a phenomenon that has not been incorporated into any current ^{222}Rn flux estimates.

During the planning of the project, it was decided that any ^{222}Rn flux maps produced need to be "calibrated" from existing data of ^{222}Rn flux. However, since there are only a few published results of direct measurements of ^{222}Rn flux, another aspect that needed to be addressed concerned the overall reproducibility of direct ^{222}Rn flux measurements. Towards this end, a rigorous assessment of the reproducibility of ^{222}Rn flux measurements using the "accumulation can" technique was investigated through field studies. These results were presented at the American Geophysical Union's 1994 Fall Meeting in San Francisco, CA. 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 ~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. This question was also addressed at the Sixth International Radon Metrology Program Intercomparison Test and Workshop organized and held at EML from April 28 to May 10, 1995 (see Summary No. 3.4).

During this exercise, researchers from around the world intercompared their measurements of ^{222}Rn flux from a standard concrete slab, as well as in the field. The results showed that the researchers agreed to within ~ 25% of each other in the field and to within ~ 15% of each other in the laboratory-based standard slab measurements (Hutter and Knutson, 1996). These results provide a basis from which to evaluate the feasibility of collectively using ^{222}Rn flux data collected by many different researchers using a variety of techniques in calibrating any global ^{222}Rn emission rate database.

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EOS, 76, S76 (1995)

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"Results of the Sixth International Radon Metrology Program Intercomparison Test and Workshop: The State of the Art in Measuring Soil Gas Radon and Radon Exhalation From Soil"
IAEA TECDOC, in press

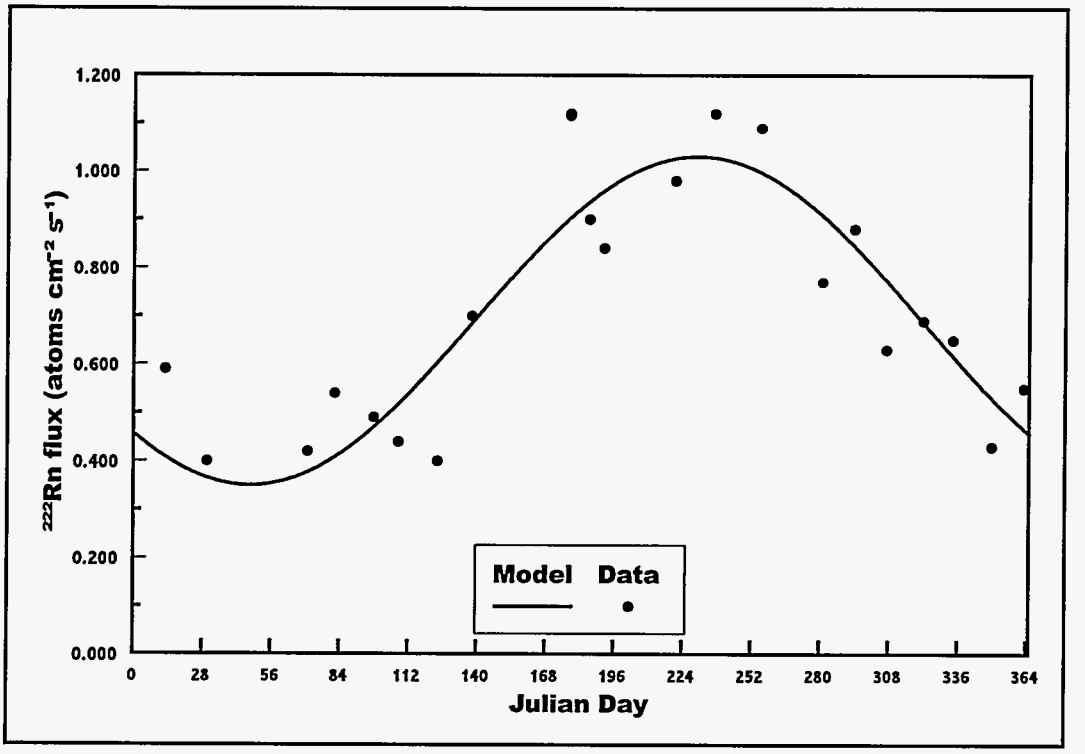

Figure 5.9 Results of ^{222}Rn flux data and a model based on a data fit following a Fourier analysis. Each data point represents the arithmetic mean of 16 individual measurements. Uncertainties associated with the data points are calculated from the standard deviation/the arithmetic mean for each data point, the arithmetic mean for all the points being ~ 20%. The correlation coefficient (r) between the data and the model is 0.90.

6 METROLOGY, CONSULTATION, AND EMERGENCY RESPONSE

6.1 OVERVIEW

Harold L. Beck

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 Technical Services 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 in which we participated during 1995 and which continued to include important interactions with the Nuclear Regulatory Commission (NRC) are also discussed in this section. There were no significant emergency response activities during 1995.

6.2 INSTRUMENTATION DESIGN, DEVELOPMENT AND SUPPORT

Technical Services Division Staff

The Technical Services Division continued to provide design and support services for EML field and laboratory projects during 1995. 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, only a few of the more significant efforts are listed below.

ARM Site Aerosol Sampling: The "statement of work" document, which was prepared at the Laboratory and includes detailed drawings and procedures to facilitate installation, received accolades from the ARM site personnel as among the best they have seen. Because of this, the installation of the articulating sampling stack and manifold, which was performed by a contractor, went very smoothly. To complete the testing initiated last year, Technical Services personnel modified commercial instruments, designed valve controls and interfacing circuitry and also completed the wiring and cabling along with the layout of the power busses. Once the complex array of instruments was tested, it was disassembled and shipped to the ARM site at Lamont, OK, where it was installed by EML personnel (see Summary No. 4.2.)

Atmospheric Chemistry Program (ACP): This program, requiring several months of effort, involved the design and construction of three instrument packages: an automatic radon gas sampler, the "Radgrabber", and the aerosol measuring system (see Summary Nos. 5.3 - 5.5). These were successfully flown on the DOE G-1 aircraft this past summer. To facilitate this installation, a plywood mockup of the G-1 cabin was constructed to exactly locate the window probes as well as the floor mountings. This effort was rewarded when EML personnel were able to complete the installation of all three instrument packages in much less time than other project participants.

Remote Atmospheric Measurements Program (RAMP): The major effort in this program area was the construction of an Automatic Remote Atmospheric Measurements System (AUTORAMS) which was completed and successfully deployed at McClellan Air Force Base, Sacramento, CA (see Summary No. 1.4). In addition to designing the air sampler, counting shield, and gripping jaws, many corrections and modifications had to be made to the pick-and-place gantry which was supplied by a commercial vendor. These included new position sensors and extensive rewiring to install all the moving cables in movable wireways (looms) using special cables resistant to continuous flexing. Other major assemblies included the design and construction of the power supply, power management unit, computer interface, limit and crash switches, and diagnostic indicators. An operation and installation manual for the AUTORAMS was written for use by the test site personnel.

Other RAMP-related work included the installation of a new system at Pinedale, Wyoming, evaluation of new notebook computers, and construction of systems capable of both modem and/or satellite transmission. Support for other RAMP locations continued, with questions and difficulties resolved via FAX, E-mail, letters or phone. RAMPSCAN, a portable gamma radiation measurement and analysis system, was built and delivered. This unit is used to assay freshly collected aircraft filters to determine the likelihood of fission products having been collected. The entire RAMPSCAN is contained in a small suitcase and can operate on its rechargeable batteries for over four hours.

Electronic Development: Many of the instruments developed at EML require the "Tattletale" computer along with sensors. Rather than perform a custom layout each time, a general purpose unit was designed and built. This unit provides the circuitry for pressure, relative humidity, and temperature sensors. Also included is an analog expansion circuit which provides up to 30 analog channels. This is a low power design and is compatible with battery operation. Memory expansion is also provided.

Environmental Science Division Support: A TLD calibration stage was designed and built, and design work on a 4-meter long shadow calibration table was initiated. Additional modifications were made to the portable TLD reader described in last year's annual report (Azziz et al., 1995). For the high altitude neutron spectroscopy program, in addition to preparing the instrumentation, a multichannel temperature instrument was designed and built for the scintillation detectors. A portable tripod on wheels for making field measurements with liquid nitrogen-cooled Ge detectors was tested at a fixed dewar height. When complete, this assembly will have the capability of adjusting the dewar height.

Radon Detection Instrumentation Development: Construction of the Barrel Radometer (Negro et al., 1995) was completed. Tests with room air indicate that the unit is functional. However, determining the actual calibration requires further work in our standard radon chamber. Some additional testing on the air conditioner which controls the dew point is also required.

Some additional work was done on the flow-through Ionization Chamber Detector. A grid was added to indicate the presence of a desired alpha decay. This allows coincidence counting which improves noise rejection. However, the spectroscopy resolution obtained thus far has not been adequate and additional development is required.

General: The EML machine shop was upgraded with the addition of a large-bed heavy duty drill press. Our printed circuit Computer-Aided Design (CAD) facility was upgraded with the installation of PCAD version 8.0. Our technicians and machinists completed a total of well over 1000 work orders during 1995.

Other maintenance activities and/or improvement of Laboratory experimental equipment during 1995 included: (1) the completion of another shield for the Quality Assurance and Metrology Division's germanium diode gamma-ray counting facility; (2) expansion of the Laboratory's Local Area Network (LAN); (3) maintenance of the alarm system for the high-security room; (4) maintenance and repair of the Laboratory's perimeter alarm/smoke system, the 60 KVA uninterruptible power system for the EML computer facility, and the electron microscope facility. The staff also continued to maintain and support the EML Chester, NJ, regional baseline monitoring station.

References

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USDOE Report EML-571 (1995)

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"Barrel Radometer"
in: "EML 1994 Annual Report"
USDOE Report EML-571 (1995)

6.3 LABORATORY COMPUTER CENTER

Camille G. Marinetti, Robert Stocco and Jenny May

Facility Enhancements: An INTERNET connection including e-mail and World Wide Web capability was added to the Laboratory's computer facility, and a Sealeze 42C poster size laminator was added to the computer graphics equipment.

Computer Graphics: The computer graphics facility is available to EML personnel for the in-house production of slides, overheads, posters and high-quality prints. Users can create their own graphics on their IBM compatible or MacIntosh PCs, or give the Computer Group 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. 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. All printed output can be laminated if necessary.

DEC Pathworks LAN: The VAX cluster with the DEC VAX 6310 and 6410 computers supports approximately 80 users via an ETHERNET LAN and a system of servers throughout the Laboratory. PC users can communicate and share resources among the VAX, IBM compatibles, and MacIntosh computers using the DECNET protocol running over ETHERNET thin wire coaxial cables connected to the ETHERNET thick wire spine which runs around the Laboratory.

The DEC Pathworks PC LAN allows PC users to share files, printers, and software between network PCs and the VAX cluster. 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 which are available through the network. PC software currently installed on the LAN includes WordPerfect (DOS and Windows), Microsoft Word, PowerPoint, Harvard Graphics for Windows, Excel, Quattro Pro, Microsoft Access, FoxPro, Paradox for Windows, and Word for Word. A LAN-based backup package allows automated backups of PCs over the network during off-hours.

INTERNET Connection: The Laboratory's LAN is now connected to the INTERNET through a "firewall" server. The server provides e-mail and allows outward-bound World Wide Web, FTP, and TELNET access for the PCs connected to the Laboratory's LAN, while preventing unauthorized access from outside. It also provides an FTP site for the Laboratory (**eml.doe.gov**).

Most of the network PCs have been set up for INTERNET access, including the installation of the TCP/IP network protocol and an e-mail program, either Eudora or Pegasus. All employees also have access to World Wide Web browsers and FTP and TELNET programs for both Windows and MacIntosh.

The Laboratory has a World Wide Web server connected outside the firewall which provides access to the new EML Web site (**www.eml.doe.gov**) for information about major EML programs, downloading EML publications, and a searchable interface to the FY95 DOE Human Subjects Research Database.

PC Repository:

The PC repository is maintained as a central pool of used computer hardware and software. When employees contribute items which they are no longer using, the item is added to the repository if it is in working order and can be of use to another person or if it is potentially valuable for replacement parts. The items are then distributed as needed.

6.4 DEVELOPMENT AND MAINTENANCE OF THE DEPARTMENT OF ENERGY HUMAN SUBJECTS RESEARCH DATABASE

Camille G. Marinetti and Lambros Kromidas

The Department of Energy (DOE) Human Subjects Research Database (HSD) contains information relating to research projects involving human subjects. These projects are funded by the Department of Energy, or are performed at DOE facilities with support from other sponsors, or performed by DOE personnel, or DOE contractor personnel. Research involving human subjects includes a variety of activities ranging from actual experimentation to simple questionnaires. The inception of this database resulted from the Secretary of Energy's "Openness Initiative" which declared that information about research involving humans must be made accessible to the public.

A computer program using Microsoft FoxPro was developed for collecting data on the use of human subjects in research at different DOE sites (see Summary No. 6.5). Both a hard copy questionnaire and computer diskettes, in Windows or MacIntosh format, were distributed as well as instructions and explanations to assist the investigators in completing the questionnaire. The information solicited relates to site information, funding, Internal Review Board (IRB) approval, where the study was conducted, who the investigators were, the number of human subjects involved in the study, description of the project, information on funding, and associated risks to the human volunteers.

After electronic completion of the questionnaire, the data files were copied onto a diskette. This diskette, along with a hard copy, was sent back to EML for proof reading and quality assurance. Subsequently, the information was incorporated into a database and printed in Hypertext Markup Language (HTML) for use on the World Wide Web. Finally, Common Gateway Interface (CGI) programs were written to allow World Wide Web users to search the database for keywords in the text portions of the project data or for specific types of use of human subjects (e.g., exposure to radiation, sampling of tissues, questionnaires, etc.). The database searches listings of all sites and accesses project information through the INTERNET address:

http://www.er.doe.gov/production/oher/humsubj/database.html.

Future plans include improvement of the data entry program to increase reliability and flexibility, to improve printouts, and to provide the ability to have sites enter data directly on-line through the World Wide Web.

6.5 FY 95 HUMAN SUBJECTS RESEARCH DATABASE

Lambros Kromidas, Camille G. Marinetti, and Merrill Heit

A searchable database was developed for DOE (see Summary No. 6.4) to describe research projects using volunteers. The database can be accessed in the World Wide Web under:

http://www.eml.doe.gov/hsr95/hsr1995.htm.

The DOE's FY 95 (October 1, 1994 - September 30, 1995) Human Subjects Research Database (HSD) contains information relating only to research projects involving humans that are funded by the DOE, performed at DOE research facilities, performed by DOE personnel, and/or by DOE contractors. The HSD contains detailed descriptions of each research study and provides information related to funding, where the study was conducted, who the investigators were, the number of human subjects involved in the study, description of the project, and associated risks, if any , to the human volunteers. The data is also presented in an abridged form for quick referencing. In addition, the database contains a glossary of scientific terms used to describe the research.

Some of the projects profiled in the HSD are therapeutic in nature; some are efforts to develop new instrumentation or techniques; some involve the use of trace quantities of radioactive material in imaging studies; others involve only the analysis of blood or urine samples from volunteers; and still others involve followup studies on workers previously employed at sites that stored or used radioactive materials. Other research projects are epidemiologic in nature, involving only the analysis of medical records of subjects to identify patterns of illness. As a result of reviewing thousands of anonymous records, a relatively large number of human subjects was added to the DOE HSD. To ensure that the database remains current, it will be updated regularly, both to add new information and to correct existing data if warranted.

Some general contents in the FY 95 HSD are:

- There are 35 reporting research facilities in the current database. Fourteen of these research facilities are DOE laboratories and 21 are non-DOE laboratories (such as Hospitals and Universities). Of the 35 reporting research facilities only one is located outside the United States and that one is in Italy (Azienda Ospedaliera Policlinico di Modena). The combined research facilities reported 225 projects, totaling $37,088,507. The number of human subjects that participated in these studies was approximately 30,858. Because epidemiological studies were included in this database, the total number of human subjects reported is not representative of actual people, but includes a large number of records from human subjects. In fact, 27,577 human subjects were part of 81 research projects that relate to epidemiological studies. The budget for these studies was $16,875,225. The 21 non-DOE facilities reported 28 projects with a budget of $11,522,138 and the participation of 4,711 human volunteers.

- Sixty-three of the 225 projects listed in this database were not supported by DOE grants. The reason these projects were listed in the DOE's Human Subjects Research Database was because the research was performed in DOE laboratories or by DOE employees. The budget for these 63 projects was approximately $8,955,662 and they included 6,658 human volunteers.

- Twenty-two research projects reported this year received support from both DOE and non-DOE grants. The budget for these projects was approximately $9,193,557. The participation of human volunteers in these research projects was 1,700.

- Of the 35 research facilities listed in this database, Brookhaven National Laboratory reported the most projects. They reported 46 research projects with a budget of $4,472,500 and 856 human volunteers.

- Of the 225 projects listed in this database, approximately 64 involved human exposure to radiation. The nature of the exposure was therapeutic, clinical, or occupational. The number of human volunteers participating in these studies was 4,351 with a budget of $11,625,108. In all the studies, radiation was used either as a tracer to facilitate imaging, or for diagnostic and/or therapeutic reasons. DOE does **NOT** support any research in which the effect of radiation upon humans per se is the object of study.

6.6 ACTINIDE METROLOGY: CHROMATOGRAPHIC RESINS FOR THE ISOLATION OF ACTINIDES

Anna Berne

A new method for determination of americium in soil aliquots up to 50 g was presented at the Eichrom Industries, Inc. Annual User's Workshop held at the 41st Conference on Bioassay, Analytical and Environmental Radiochemistry on November 13, 1995 in Boston, MA. This method will also be presented at another User's Workshop in March 1996 in Belgium. It will also be publicized through an Eichrom Newsletter and eventually will be submitted for publication in a peer-reviewed journal.

The new method (Figure 6.1) combines the classical means of sample purification required by the larger aliquots of soil with time- and waste-reducing use of Eichrom Resin extraction columns. It is estimated that for a batch of four to eight samples, the number of man-hours for the americium analysis can be as little as 6.5 h. The total time for the determination is about four days, that is from the first oxalate co-precipitation to the point of submission for alpha-spectrometry. Table 6.1 compares results for determination of americium using this method with those obtained using the Am-06 Procedure (Berne and Greenlaw, in press).

Comment: The reported uncertainty is the standard deviation of the average. The individual determinations are based on enough counts so that the combined counting error (for the tracer and the analyte) is never more than 7% of the calculated value.

Reference

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

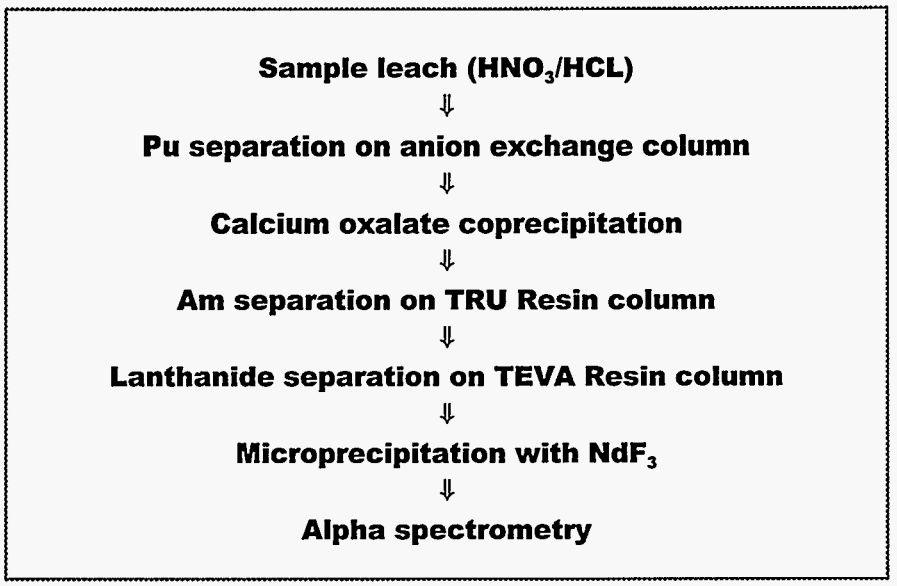
TABLE 6.1

SUMMARY OF THE RESULTS OF ^{241}Am DETERMINATION IN SOIL ALIQUOTS UP TO 50 g

Sample size (grams)	mg Ca present	recovery of spike (%)	^{241}Am Bq kg^{-1}	$\bar{x}$ new method Bq kg^{-1}	$\bar{x}$ Am-06 procedure Bq kg^{-1}
Japanese ref. fallout material					
4	350	87	2.13		
4	350	81	2.37	2.36 ± 0.14	
4	350	70	2.40	(± 6.0%)	N/A
4	350	87	2.52		
4	350	82	2.38		
QAP 9309-Soil					
	110	70	0.24	0.234	0.25
30	110	60	0.23	± 0.006	
30				(± 2.7%)	
QAP 9509-Soil					
	200 + N/A	87	1.95	1.93 ± 0.18	1.79 ± 0.07
20	200 + N/A	73	1.75	(± 9.4%)	(± 4%)
35	200 + N/A	61	2.11		
50					

NOTE: Japanese reference material available from EML's participation in an intercomparison exercise with the Meteorological Research Institute in Japan.

a) New method:



b) Am-06 procedure:

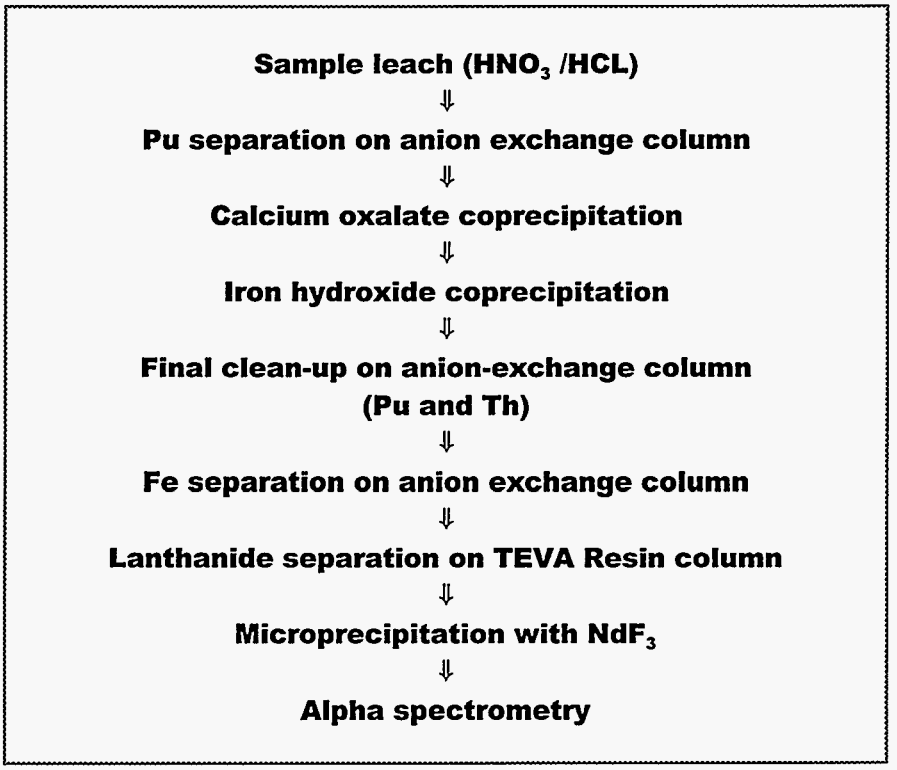


Figure 6.1 Flow Chart for Americium Separation in Soil.

6.7 RAPID DETERMINATION OF RADIUM BY SOLVENT EXTRACTION AND PERALS™*

Salvatore C. Scarpitta

The extraction efficiencies (EE's) of ^{226}Ra and its progeny ^{210}Pb/Po have been determined using a commercially available toluene based extractant under various chemical conditions. Six potentially interfering α emitting actinides (^{232}U, ^{230}Th, ^{244}Cm, ^{242}Pu, ^{243}Am and ^{237}Np) were also tested. Radium can be directly extracted from an aqueous sample with a 1-2 mL aliquot of RADAEX^{R} and measured in a very low background PERALS™ α counting system that electronically rejects unwanted γ / ß signals. The extractant can also be mixed into either a toluene based or water miscible Liquid Scintillation (LS) cocktail and measured by conventional LS counting for rapid screening purposes. The EE of ^{226}Ra into RADAEX was 98 - 100% from a 0.3M $NaNO_3$ solution adjusted to pH > 10, whereas that of ^{210}Pb/Po was 2-3%. The EE's for ^{226}Ra, ^{210}Pb/Po and the 6 actinides were also measured over a pH range of 1 to 12. Of the 6 actinides, only Cm and Am co-extracted at pH > 10 with EE's of 100% and 63%, respectively. These 2 actinides, if present in a sample, cannot be back-extracted from the Ra bearing organic phase by simply changing the pH or acid concentration without loss of Ra isotopes. Americium or Cm, if present, would interfere with the determinations of either α emitting ^{223}Ra (5.6 MeV), ^{224}Ra (5.7 MeV) or ß emitting ^{228}Ra (via ^{228}Th progeny; 5.3 MeV α) but not that of ^{226}Ra (4.8 MeV α) because of the 0.250 MeV α energy resolution of the PERALS™ Spectrometer. Gamma emitting ^{133}Ba, which may be used as a yield determinant, is not detected by PERALS™. This work was presented at the 29th Midyear Health Physics NORM (Naturally Occurring Radioactive Materials) Topical Meeting, in Scottsdale, AZ, in January 1996.

* Ordela, Inc., Oak Ridge, Tennessee

6.8 SOLID PHASE SCINTILLATION COUNTING OF α, β AND γ EMITTING RADIONUCLIDES USING PLASTIC SCINTILLATING POLYMER BEADS

Salvatore C. Scarpitta and John Kada

As a waste minimization alternative, gram amounts of two commercially available plastic Beaded Scintillation Materials (BSMs) are being investigated and compared to a commercially available water-miscible liquid scintillant using selected NIST traceable α, ß and γ emitters. These beaded polymers can be used in selected applications to significantly reduce the mixed-organic liquid waste generated from routine Liquid Scintillation (LS) counting of aqueous environmental samples. Conventional LS counting requires 5-20 mL of LS cocktail per sample, producing a non-reusable mixed-waste end product.

The first of the 2 BSMs, a polyvinyltoluene (PVT) polymer* with 100-250 ∝m diameter, produces 425 nm photons that are detected by a Packard Tri-Carb 2250-CA LS Analyzer# with 90-94% counting efficiency (CE) when 5 or 10 gm of the dry polymer was mixed with 1 gm ^{40}KCl (1.320 MeV $ß_{max}$ as a chloride salt; 851 dpm ^{40}K g^{-1} KCl) in either 20 mL glass or plastic counting

vials. The CE dropped to 70% when 0.5 gm of the dry material was used. When water was added to the ^{40}KCl/BSM mix, the CE ranged from 60-90%, depending on the weight ratio mix of BSM to water. An 84% CE was obtained when 2 gm of the PVT material was slurred with 1 mL of the KCl solution.

The second BSM (of comparable size and density) was a polyvinyl benzene (PVB) polymer containing 5% divinylbenzene as an additive. It yielded a maximum CE of 72% when 0.5 - 1.0 gm of the dry material was mixed with 1 gm ^{40}KCl. The CE was reduced by a factor of 2 when 10 gm of the dry PVB material was mixed with ^{40}KCl. As with the PVT material, the optimum mix of polymer to KCl solution was 2:1 for the PVB material.

The relationship between CE and beta energy was linear for both BSM's using 5 pure ß emitters whose E_{max} ranged from 0.245 MeV (^{45}Ca) to 3.50 MeV (^{106}Ru). Using 2 gm of PVT beads with 1 mL of the aqueous solution of the nuclides, the CE increased from 22% (for ^{45}Ca)) to 100% (for ^{106}Ru). Using ^{244}Cm (5.2 MeV) as an α source, the α-CE for the dry PVT material was 65%, whereas that of the dry PVB material was 95%. When washed with water, neither material retained any previously added α or ß activity.

* Bicron Corp., 12345 Kinsman Rd, Newberry, OH. 44065
# Packard Instrument Co. Downers Grove, IL.
+ Duke Scientific Corp. 2463 Faber Pl., PO Box 50005, Palo Alto, CA. 94303.

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Int. J. Appl. Rad. Isotopes, in press

6.9 PLASMA SPECTROMETER UPGRADE

William C. Rosa

Our plasma spectrometer capability was upgraded via the integration of several pieces of new hardware. Installation of an Analytical Data Manager and an Ultrasonic Nebulizer was completed. The preliminary results of these improvements to the plasma spectrometer are: (1) an increased data collection capability by a factor of 10; (2) an improved signal-to-noise ratio by a factor of 5 for many of the analytes of interest; and (3) a substantial reduction in use of consumables, for example argon gas. These improvements will decrease both sample analysis time and operating costs.

6.10 GAS CHROMATOGRAPHY/MASS SPECTROMETRY

Raymond J. Lagomarsino

A KRATOS high resolution magnetic sector gas chromatograph/mass spectrometer has been installed in a temperature and humidity controlled facility. The analyzer is a double focusing Nier-Johnson geometry, consisting of a 90° electrostatic analyzer with a mean radius of 135 mm and a 60° magnetic analyzer with a mean radius of 205 mm. Operable range is from 2∝ to 4000∝. Source capabilities include electron impact and positive/negative chemical ionization. Instrument control, acquisition and data processing is by a Sun Microsystem SPARC-station computer (UNIX).

The system is being evaluated for the measurement of volatile organic compounds in performance evaluation samples for DOE's Mixed Analyte Performance Evaluation Program (MAPEP). Applicability of the system for EPA Method 8260A will be investigated. A HP-7695 Purge and Trap System is available for installation once the acceptance criteria have been met.

6.11 GAMMA-RAY SPECTROMETRY

Colin G. Sanderson, William Rivera, 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 nine high resolution germanium detectors in five different configurations. These consist of three 1.5 cm diameter wells, a 2.54 cm well, a 1.5 cm well in a segmented germanium detector with a sodium iodide Compton suppression shield, four 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 cubic centimeter Marinelli (reentrant) beakers, 90 cubic centimeter aluminum cans, 45 cm^3 plastic planchettes and 1 to 2 cm^3 samples in plastic test tubes for well counting.

During the past year, 924 filter samples from the Environmental Science Division's Surface Air Sampling Program (see Summary No. 1.3) were routinely analyzed with germanium detectors for ^{7}Be, ^{95}Zr, ^{137}Cs, ^{144}Ce and ^{210}Pb. In addition, over 1231 QAP samples, intercomparisons and misc-ellaneous samples were analyzed for a variety of gamma-ray emitters using these systems.

6.12 *IN SITU* GAMMA-RAY SPECTROMETRY

Kevin M. Miller, Peter Shebell, Marcel Reginatto and Gladys A. Klemic

The technique of gamma-ray spectrometry as applied directly in the field for assessing environmental radiation and radioactivity has been refined and applied to many studies by EML researchers over the course of four decades. Since the technique has the potential to expedite radiological surveys in support of environmental restoration, it is receiving increased attention. This past year, we provided consultation to 22 different organizations seeking guidance on performing *in situ* spectrometric measurements for site characterization and remediation control. In the interest of further spreading the use of this technology, we organized and taught a short course on *in situ* gamma-ray spectrometry for the IEEE Nuclear Science Symposium held in San Francisco in October. This was the fourth time in the past five years we have given this course at the request of the IEEE. In this time span, close to 100 individuals from various private companies and government and international organizations have taken this course.

In 1995, EML was under contract to the Nuclear Regulatory Commission to provide guidance and support for applying *in situ* spectrometry to measuring residual radioactivity during decommissioning surveys. We demonstrated the technique during two surveys and also provided a one-day training program (see Summary No. 6.20). Demonstration to DOE contractors was also provided at the Weldon Spring Remedial Action Site (see Summary No. 6.13).

The International Commission on Radiation Units and Measurements (ICRU) issued its report on *in situ* gamma-ray spectrometry in 1995. This work was the collaborative effort of a six member committee with individuals from five different countries, including an EML scientist who represented the U.S. The committee began its work in 1991 and final editing and review of the report was completed in 1994 (ICRU, 1994). This report will serve as a primer for the beginner and as a general reference for the expert field spectroscopist. Also, as an international consensus report, it will provide impetus in having this measurement technique gain wider acceptance in the regulatory community.

Reference

International Commission on Radiations Units and Measurements
"Gamma-Ray Spectrometry in the Environment"
ICRU Report 53 (1994)

6.13 DEMONSTRATION OF *IN SITU* GAMMA-RAY SPECTROMETRY AT WELDON SPRING SITE REMEDIAL ACTION PROJECT

Peter Shebell and Kevin M. Miller

The Weldon Spring Site Remedial Action Project (WSSRAP) in Weldon Spring, Missouri, requested an on-site demonstration of *in situ* gamma-ray spectrometry. WSSRAP has several possible applications for *in situ* gamma-ray spectrometry for determining concentrations of uranium and thorium in surface soils. These applications range from excavation control during soil remediation to a reduction in the number of conventional confirmation samples. WSSRAP staff was also interested in the technique for use in addressing areas that are not amenable to standard sampling techniques. EML provided DOE personnel and contractors with an overview of *in situ* gamma-ray spectrometry and a demonstration of the technique along with an interpretation of the *in situ* results as they compare to traditional soil sampling methods. (See Figure 6.2).

Three sites were used for the demonstration. One site was believed to be at or near background levels for uranium and thorium, while the other was above background but below the action level. The third site was a wooded area several miles from the facility known as the southeast (SE) drainage ditch. With the exception of the drainage ditch, the measurements were performed on a grid which approximated the sampling grid described in WSSRAP's cleanup confirmation plan (USDOE, 1994). Each grid consisted of five measurement points, one at each corner of a 20 m square with one at the center. Measurements at the SE drainage ditch were taken at points believed to be contaminated with thorium.

In addition to the standard 1-meter measurements, a series of shielded measurements was performed at about 15 cm from the ground. It was anticipated that these "point" measurements would replace a significant fraction of conventional confirmation samples. A total of 20 (shielded and unshielded) *in situ* spectra was collected and analyzed over the course of three days. Measurements of the total exposure rate from penetrating radiation (cosmic and terrestrial) were also made at the grid points with a pressurized ionization chamber. These measurements serve as a component of quality assurance for the 1-meter measurements. The demonstration concluded with a presentation of preliminary results. The *in situ* results of uranium concentrations were consistent with previous measurements taken at or near the measurement locations. Subsequent soil samples taken at the measurement locations show good agreement with *in situ* results for uranium. WSSRAP is currently considering the technique to reduce the number of conventional confirmation samples.

Reference

U.S. Department of Energy,
Chemical Plant Area Cleanup Attainment Confirmation Plan Rev. 0.
Contract No. DE-AC05-86OR21548. Weldon Springs Site Remedial Action Project. Weldon Spring, Missouri. November 1994.

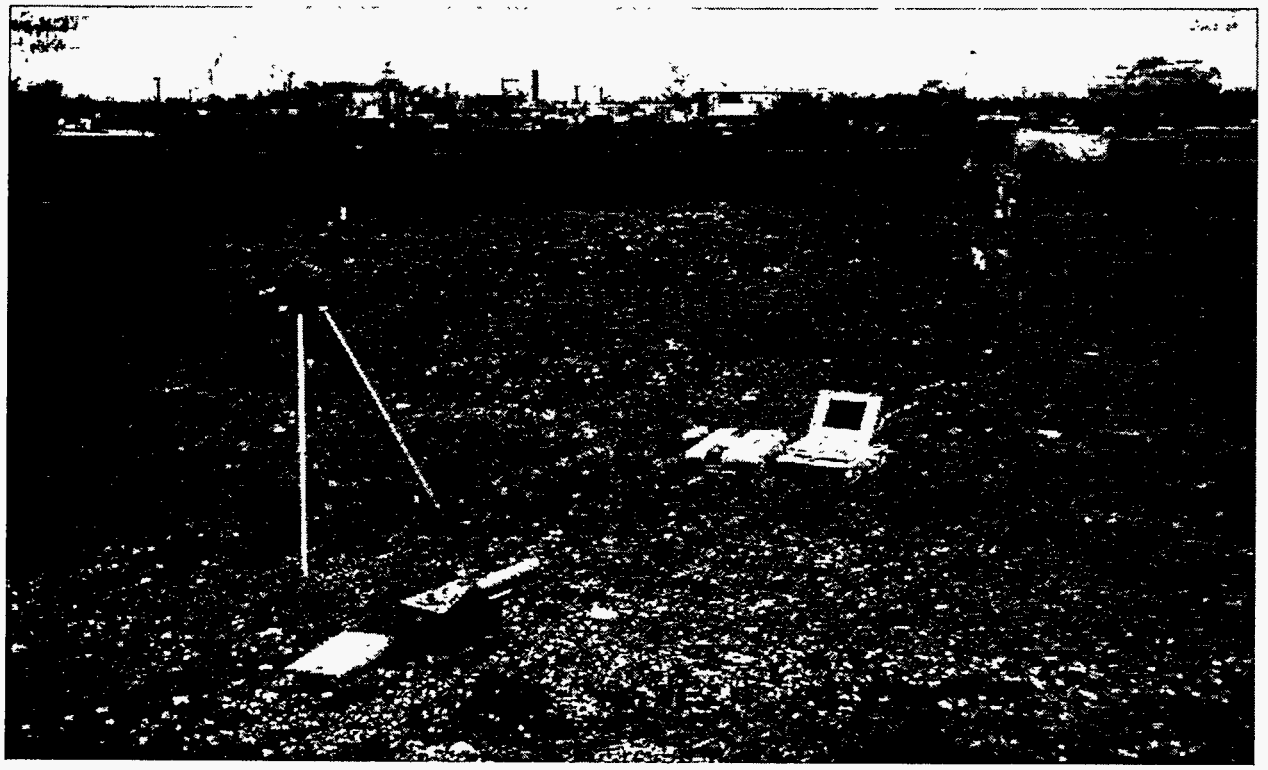

Figure 6.2 An in-situ gamma-ray measurement at the Weldon Spring Site Remedial Action Project (WSSRAP) in Weldon Spring, Missouri.

6.14 ANALYSIS OF *IN SITU* MEASUREMENTS ON A GRID

Marcel Reginatto, Peter Shebell and Kevin M. Miller

In situ spectrometric measurements can be performed on a square or triangular grid to ensure complete coverage of extended areas that are potentially contaminated with radionuclides. While such measurements will provide valuable information on the spatial distribution of the contaminant, the problem of reconstructing the real distribution of activity on the ground from a finite number of measurements is ill posed and the solution is not unique. Difficulties also arise due to the experimental errors associated with the set of measurements.

An important element in surveys of residual radioactivity is the detection of localized areas of elevated contamination, sometimes referred to as "hot spots". We have developed a computer code that analyzes a series of *in situ* measurements on a grid and searches for distributions of activity (possibly many) that fit the data and on which a hot spot is present. The algorithm makes use of a preliminary deconvolution of the data using the maximum entropy method (Skilling and Bryan, 1984), followed by further analysis which results in solutions each with the largest possible hot spot at a given location over a smooth background. The algorithm is quite general and could be modified for use in other types of measurements. The computer code is designed to be used as a tool when evaluating compliance with regulations that set limits on hot spots and is, therefore, designed to identify those "worst case scenarios" where a hot spot might be present.

Properties of the algorithm were investigated using simulated data as well as actual measurements. In one field test, a 24.05×10^3 γ/s Cs-137 check source was used to simulate a hot spot, and 35 equally spaced measurements were taken on a rectangular 7m x 5m grid with measurement points 2.44 m apart. Figure 6.3 shows the output from the preliminary maximum entropy deconvolution, while Figure 6.4 shows the final surface activity distribution that corresponds to the largest single potential hot spot over a smooth background (both are good solutions in the sense that they both fit the data). While the maximum entropy solution shows structure that reflects the grid geometry (with the peaks centered exactly at measurement points where there is an excess of activity), the solution with the largest hot spot has only one large peak that falls between measurement points. The code placed the largest potential hot spot within 0.67 m of the actual location of the check source. In another successful application, the code was used to analyze measurements on a triangular grid at a facility where depleted uranium was used in industrial operations. This last application is related to Nuclear Regulatory Commission decommissioning criteria work in progress at EML (see Summary No. 6.20).

Part of this work was presented at the IEEE 1995 Nuclear Science Symposium and Medical Imaging Conference and a paper was submitted for publication (Reginatto et al., in press).

References

M.Reginatto, P.Shebell and K.M.Miller
"An Application of the Maximum Entropy Method for Assessments of Residual Radioactivity at Contaminated Sites"
IEEE Transactions on Nuclear Science, in press.

J.Skilling and R.K.Bryan
"Maximum Entropy Image Reconstruction: General Algorithm"
Mon. Not. R. Astr. Soc., 211, 111-124 (1984)

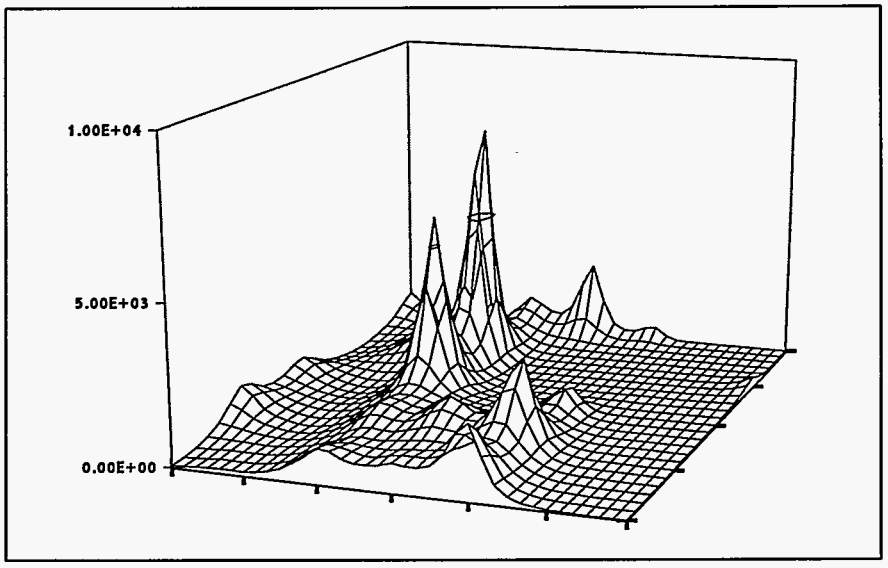

Figure 6.3 Surface activity distribution ($\gamma/s/m^2$) from the maximum entropy deconvolution of the data

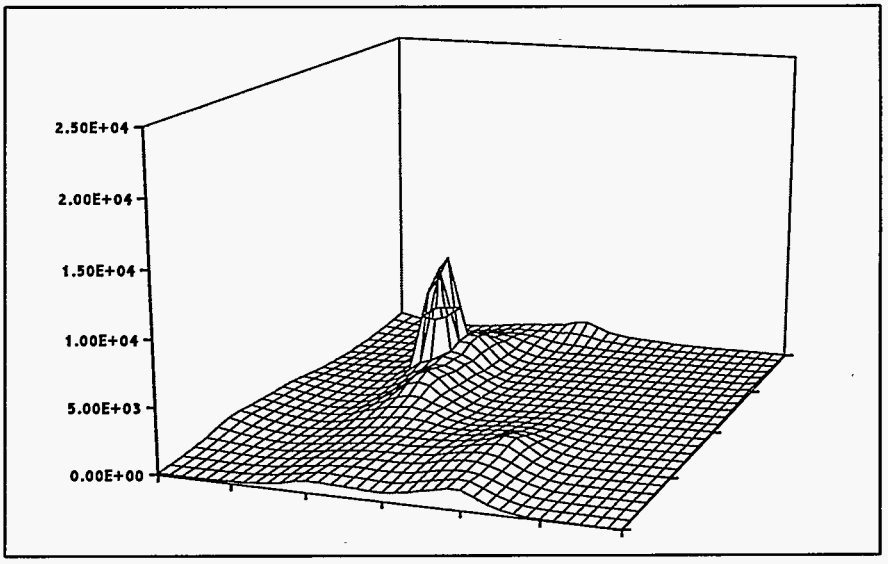

Figure 6.4 Surface activity distribution ($\gamma/s/m^2$) corresponding to the largest potential hot spot over a smooth background.

6.15 SEMI-EMPIRICAL METHOD FOR DETERMINING THE ANGULAR RESPONSE FOR Ge DETECTORS

Peter Shebell and Marcel Reginatto

The calibration of a coaxial Ge detector for *in situ* spectrometry involves the determination of its angular response. The current EML procedure to measure this quantity is simple, but time consuming. It requires a series of measurements which are fitted to a polynomial for a few calibration energies. Spurious results can occur when poor statistics for weak gamma emitters cause fluctuations in the data which are incorporated into the polynomial fits. Subsequent interpolation and extrapolation of the angular response for a given energy is difficult and subject to error.

A general procedure for determining the angular response for coaxial Ge detectors is being developed which incorporates the basic physics of photon interactions within the active region of the crystal. The approach is based on the work of Moens et al. (1981) and Wang et al. (1995) in connection with the calculation of the absolute-peak-efficiency for HPGe detectors for various counting geometries. Their semi-empirical method involves the measurement of the efficiency of the detector for a reference geometry as a function of energy and the calculation of an effective solid angle for an experimental reference configuration. The efficiency for a particular configuration is then expressed in terms of the ratio of effective solid angles and the efficiency for a reference geometry.

In similar fashion, the angular response of a HPGe detector for an *in situ* measurement , $R(\theta)$, can be expressed as the ratio of effective areas:

$$R(\theta) = A_\theta / A_{\theta=0^\circ}$$

where A_θ is the effective area for a parallel beam of photons incident on the detector with an angle θ relative to the symmetry axis of the crystal. A_θ is defined as

$$A_\theta = \int_S (P\phi_{eff})\ \mathbf{n}\cdot d\mathbf{S}$$

where P is the probability that a photon passing through a differential surface area, $\mathbf{n}\cdot d\mathbf{S}$, will scatter incoherently in the active region of the crystal; ϕ_{eff} is the photon fluence rate adjusted for scattering by materials in the detector.

Preliminary results for a p-type closed end coaxial detector with a relative efficiency of 17.6% and a crystal length-to-diameter ratio of 0.57 indicate that this new approach will agree with measured data within 6%. While errors of this magnitude do not contribute significantly to the total uncertainty, improved computational methods should bring agreement to better than 3%.

References

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"Calculation of the Absolute Peak Efficiency of Gamma-Ray Detectors for Different Counting Geometries"
Nucl. Instrum. Meths., 187, 451-472 (1981)

Wang, T. K., et al.
"HPGe Detector Absolute-peak-efficiency Calibration by Using the ESOLAN Program"
Appl. Radiat. Isot. 46, 933-944 (1995)

6.16 EML HOSTS THE 6TH INTERNATIONAL RADON METROLOGY PROGRAM (IRMP) INTERCOMPARISON TEST AND WORKSHOP

Adam R. Hutter and Earl O. Knutson

EML was host to the 6th IRMP Intercomparison Test and Workshop titled "State of the Art in Measuring Soil Gas Radon and Radon Exhalation From Soil", June 12 - 15, 1995, the week following the Natural Radiation Environment VI Symposium. Twenty-six (26) participants representing 11 countries and 23 different institutions attended the workshop. Three different activities were planned for the week: a day of seminars and discussions, a day of laboratory measurements of radon flux and high-concentration grab samples, and a day of field measurements of radon flux and soil gas radon. The first day included nine presentations, with ensuing discussions, aimed at addressing the question of whether the current state-of-the-art in measuring soil gas radon and radon flux from soils meet the precision and accuracy requirements for applications of these measurements. The laboratory and field exercises were designed to quantify the intercomparability of different methods and instruments used by different researchers. During the laboratory day, a Ra-spiked concrete slab was used as a standard for radon flux measurements. Likewise, ^{220}Rn and ^{222}Rn grab samples were obtained from the source drum. The day of field exercises was held in an open grassy meadow at Cheesequake State Park in Matawan, NJ, where radon flux from the surface and soil gas measurements was obtained. The soils at the site are known to be fairly homogeneous, an important consideration for intercomparison exercises.

There was a general consensus among the participants that the precision and accuracy needs for a particular application for both radon flux and soil gas radon dictate the methods and instruments that should be used. There are instruments currently available, either commercially or strictly for research, to ensure proper and appropriate measurements (i.e., precision and accuracy needs) for virtually any application, if prudently chosen.

The results of the laboratory and field exercises show excellent agreement among the participants when comparing source drum and exhalation measurements, both from the slab and in the field, but soil gas measurements were scattered, with a greater than three-fold variation, largely attributed to errors introduced during sampling rather than calibration or counting errors.

A report will be published by the International Atomic Energy Agency (the sponsor for the IRMP) with a condensed version being submitted as an article to a peer-reviewed journal. There was also consensus on the importance of holding another similar intercomparison and workshop, perhaps in conjunction with the next NRE meeting. Topics suggested for the next workshop include studying the importance of soil permeability on soil gas radon measurements, as well as measurements of soil emanation. Figure 6.5 shows scientists performing radon flux measurements at the Cheesequake State Park during the 6th IRMP Intercomparison Test and Workshop. Figure 6.6 is a group photo of IRMP6 participants.

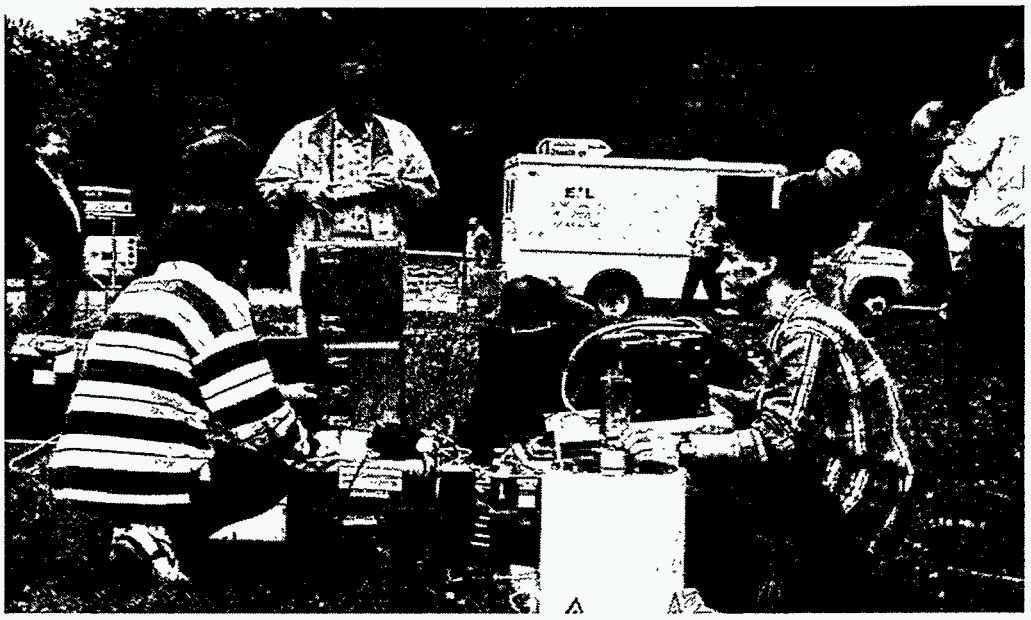

Figure 6.5 Scientists performing radon flux measurements at the Cheesequake State Park during the 6th IRMP Intercomparison Test and Workshop hosted by EML, June 12 - 15, 1995.

Figure 6.6 Group photo of IRMP6 participants.

6.17 PARTICIPATION IN THE IAEA'S MISSION TO KAZAKHSTAN

Adam R. Hutter and Peter Shebell

The final report on EML's participation in the International Atomic Energy Agency's (IAEA) Mission to Kazakhstan, "Strengthening Radiation and Nuclear Safety Infrastructures in Countries of the Former USSR, Special Task - Pre-assessment of the Radiological Situation in the Semipalatinsk and Western Areas of Kazakhstan", was submitted to the IAEA. During July 1994 a 10-member team, with participants from France, UK, Austria and Russia, visited sites within the Semipalatinsk Nuclear Test Site as well as several surrounding villages. Specifically, the objectives of the EML team were to apply independent methods and equipment to assess potential current radiation exposures to the population. Towards this end, the EML scientists collected *in situ* gamma-ray spectra, performed external gamma dose rate measurements using pressurized ionization chambers, and collected soil samples in order to estimate the inventory and to determine the depth distribution of radionuclides of interest. The report, "Environmental Radiation Measurements at the Former Soviet Union's Semipalatinsk Nuclear Test Site and Surrounding Villages," will be published as an EML Report and in a condensed form will be submitted for publication in a journal. Figure 6.7 shows an EML scientist performing field measurements near ground zero at the Semipalatinsk Nuclear Test Site, Kazakhstan.

Figure 6.7 EML scientist performing field measurements near ground zero at the Semipalatinsk Nuclear Test Site, Kazakhstan.

6.18 FIELD INVESTIGATION AT CHELYABINSK-65

Karen A. Stevenson

Since early 1994, EML has been participating in the joint Russian - United States subsurface contaminant transport study conducted under the auspices of the Joint Coordinating Committee for Environmental Restoration and Waste Management (JCCEM) between the DOE Office of Environmental Management (EM) and the Russian Ministry of Atomic Energy (MINATOM). Other participating U.S. laboratories have included Savannah River Laboratory (SNL), Pacific Northwest National Laboratory (PNNL) and Lawrence Berkeley Laboratory (LBL).

In September of 1994, the first joint Russian - U.S. field site investigation was conducted under the JCCEM in the territory of the Mayak Production Association, 13 km southwest of Ozersk (formerly Chelyabinsk-65) (Wollenberg et al., 1995). The purpose of this field site investigation was to examine the frontal area of a contaminated groundwater plume moving from the disposal site, Lake Karachai, toward the Mishelyak River. Activities during this first joint exercise included (1) isolation of hydrologic intervals in two wells and the production of water from these isolated intervals in order to compare isolated versus open-well sampling methods and to determine hydraulic transmissivities of the aquifers; (2) surface and soil-water sampling, accompanying radiometric measurement and subsequent chemical analyses; and (3) electrical resistivity profiling in areas of expected contrasting resistivity using a conventional dipole-dipole array survey instrument.

The EML team members, Karen Stevenson (Federal Technical Lead) and Wayne Lowder (consultant), were tasked with providing the U.S. team with both real-time and passive dosimetry and conducting *in situ* gamma-ray spectrometry measurements along the Mishelyak River. Figure 6.8 is a schematic of the field site. Locations listed as PT0 - PT7 indicate where Russian - United States comparative field site radiometric measurements were made, and locations listed as W1 - W7 indicate where surface river water grab sampling and *in situ* radiometric measurements were conducted. Also shown in Figure 6.8 are the approximate location of the Mishelyak River and other site-identifying features.

Results of the field site radiometric measurements (PT0 - PT7) indicate that the free-air dose exposure rate at this location is approximately three times higher than what would be expected from natural gamma-ray emitters. Investigation of the spectra indicate that ^{137}Cs was the most significant contributor to the excess external dose exposure at an average deposition of 126 ± 26 (1 SD) kBq m^{-2}. Likely source terms of this anthropogenic radionuclide include past weapons testing, global fallout and a 1967 incident involving a surface repository (Stevenson, 1994).

Russian laboratory results of grab water samples obtained at different locations along the Mishelyak River detected the presence of ^{60}Co at concentration levels that were less then 1.5 Bq l^{-1}, 0.1% of the present day permissible concentration level for radiation safety in Russia. As shown in Figure 6.9, the *in situ* gamma-ray water measurements conducted by the U.S. team in the Mishelyak River closely resemble a half-space geometry (2π). Calibration of the spectra, using this half-space geometry, corroborated the findings of the Russian laboratory results to within an order of magnitude.

Findings of the Russian - U.S. activities during the field site investigation, including the EML radiometric measurements, are contained in the following reports:

"Joint Russian-American Hydrogeological-Geochemical Studies on the Karachai-Mishelyak System, South Urals, Russia" (submitted to the Journal of Environmental Geology, 1996);

"Resistivity and Induced Polarization Survey at a Russian Nuclear Waste Site" (submitted to Geophysics, 1996); and

"Joint Russian-United States Radiometric and Surface Water Chemistry Measurements with the Karachai - Mishelyak System, Southern Urals, Russia" (submitted to the Journal of Environmental Radioactivity, 1996).

In July of 1996, a second joint Russian-U.S. field site investigation will be conducted in the territory of the Mayak Production Association.

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)

Wollenberg, H., C.-F. Tsang, W. Frangos, R. Soubau, W. Lowder, K. Stevenson, M. Foley, E. Drozhko, G. Romanov, Y. Glagolenko, A. Posochov, Y. Yvanov, L. Samsonova, A. Petrov, S. Ter-Saakain, N. Vasil'kova, and A. Glagolev
"A Joint Russian - American Field Test at the Chelyabinsk-65 (MAYAK) Site: Test Description and Preliminary Results"
"Management of Low-Level Waste and Remediation of Contaminated Sites and Facilities," Fifth International Conference on Radioactive Waste Management and Environmental Remediation, ICEM'95 **2**, pp. 1381-1386 (1995)

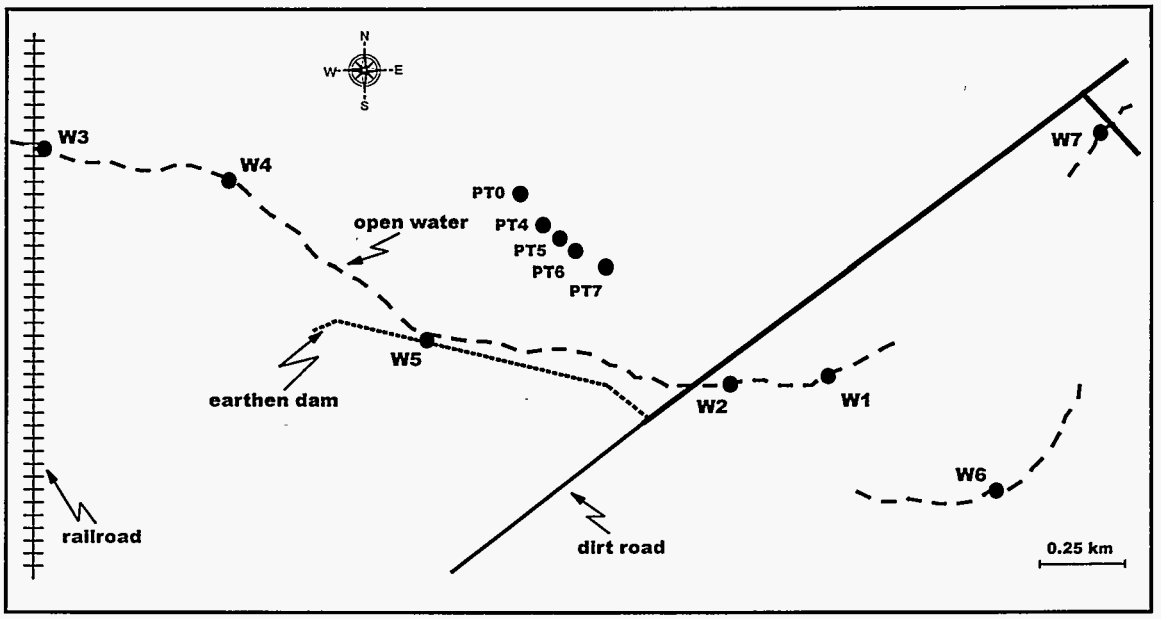

Figure 6.8 A schematic of the field site. Marked in dots (●) as PT# are locations where radiometric measurements were conducted. Marked in dots (●) as W# are locations where both radiometric measurements were conducted and water samples obtained. Also shown, are an access dirt road, a railroad tracks, an earthen dam and open water of the Mishelyak River.

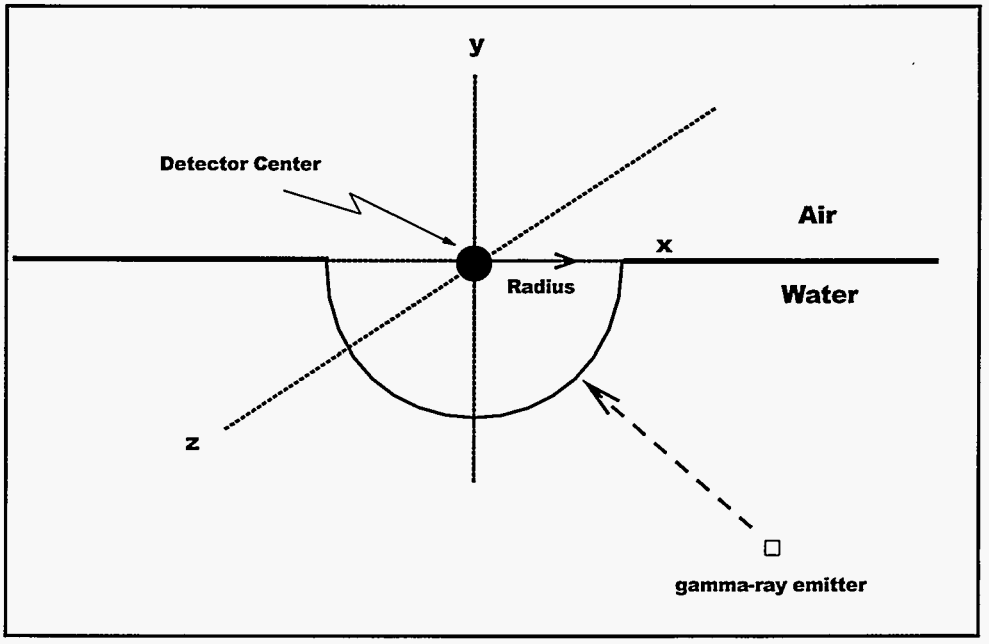

Figure 6.9 Schematic for calculating the photon fluence rate per activity in water for the *in situ* gamma-ray spectrometry measurements using a half-space geometry.

6.19 RADIOLOGICAL EVALUATION OF THE CONSTRUCTION SITE FOR THE FISSILE MATERIAL STORAGE FACILITY IN RUSSIA

Karen A. Stevenson

In June 1995, at the request of the Defense Nuclear Agency (DNA), EML participated in a radiological evaluation of the construction site for the Fissile Material Storage Facility (FMSF) located within the territory of the Mayak Production Association in Russia. The DNA requested a health and safety survey to assess the potential radiation exposure to U.S. personnel who will participate in the delivery, training and maintenance of construction equipment. Support for the construction of the FMSF at Mayak is through a collaborative project between the Department of Defense (DoD) and the Ministry of Atomic Energy for the Russian Federation (MINATOM) to build secure storage facilities for excess Russian weapons materials. United States participants for this evaluation included personnel from DNA, the U.S. Army Center for Health Promotion and Preventive Medicine, the U.S. Army Corps of Engineers and the Department of Energy's Environmental Measurements Laboratory.

A possible source of anthropogenic radionuclide contamination at the construction site was a 1967 incident at a surface radioactive waste repository, which dispersed almost 20 TBq (600 Ci) of radioactive silt over an area of 2700 km^2. At the time of the June 1995 survey, the Russians had removed the top 15 - 30 cm of soil from the construction site. A radiological survey revealed that the radiation levels do not pose any immediate danger to personnel. The exposure levels were found to be less than $< 10\ \mu R\ hr^{-1}$, with a few isolated locations exceeding $50\ \mu R\ hr^{-1}$. Recommendations of the evaluation team included: (1) a request for the Russians to remove the remaining contaminated soils at the construction site; (2) continuous air monitoring employed on the site during the time of personnel occupancy; and (3) U.S. personnel should undergo baseline bioassay performed prior to being deployed to the site and periodically thereafter (every 90 days).

6.20 SUPPORT FOR NRC DECOMMISSIONING RULEMAKING

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

Since 1993, EML has been under contract to the Nuclear Regulatory Commission (NRC) to support the implementation of a rulemaking on new decommissioning criteria. The proposed criteria for unrestricted release of a licensed facility would be a limit of 0.15 mSv (15 mrem) total effective dose equivalent per year to the average member of a critical group from residual radioactivity that is distinguishable from background. Our work initially dealt with a compilation of information on background radiation, its components and spatial and temporal variations, as well as the costs associated with making measurements of residual activity at concentration levels approaching those found in background. This provided necessary information for the preparation of the draft Generic Environmental Impact Statement to support the rulemaking.

Subsequently, EML was tasked to prepare guidance documents for the methodology to be employed in final status surveys. In 1995, the writing and editorial and technical reviews for two NUREG documents were completed and these were then published by the NRC as drafts for comment. The first document (Gogolak et al, 1995) deals with the overall final status survey design using a Data Quality Objective (DQO) approach and the requisite non-parametrical statistical tests that would be employed to judge whether an area undergoing decommissioning meets release criteria. This methodology should allow for relatively efficient survey designs compared to present draft guidance as, in general, smaller numbers of samples or measurements are required. At the same time, it provides a flexible approach in providing a defensible basis for release based on hypothesis testing and the setting of desirable type I and II error rates. The second document (Huffert and Miller, 1995) deals with the supporting measurement methodology that can be employed given the default concentrations associated with the 0.15 mSv per year dose level. Although the task of measuring residual radioactivity will be more difficult at low concentration levels, the application of nuclide specific field measurements, as with high resolution gamma-ray spectrometry, will generally provide the necessary sensitivity. The statistical and measurement methodologies were presented by EML staff at a public workshop held in September 1995 at NRC headquarters in Rockville, MD.

In addition to the methodology development, EML provided training to the NRC staff and their contractors on *in situ* spectrometric methods and non-parametrical statistical tests. As a training exercise and to further refine the methodology, demonstration surveys were performed at two facilities. One was a power reactor (see Figure 6.10) where the critical nuclides of concern were ^{60}Co and ^{137}Cs. Both an indoor and outdoor survey unit were measured using reference areas for comparison. Measurement methods employed included gross surface activity readings with a GM survey meter, total exposure rates readings using a pressurized ionization chamber, soil sampling followed by laboratory analyses, removable activity measurements using smears and *in situ* gamma-ray spectrometry. The second survey demonstration involved an outdoor survey unit at a facility where depleted uranium was used in industrial operations. In place of a reference area, the background ^{226}Ra was assessed at each measurement point to infer the "excess" ^{238}U present. Since standard scanning sensitivity was not adequate at the proposed release levels in this case, a closely spaced grid of *in situ* spectrometric measurements using a large volume Ge detector was used along with a deconvolution routine developed by EML to check for elevated area contamination (see Summary No. 6.14).

Additional development work in the decommissioning area is ongoing. This past year, one sample non-parametric tests were refined and incorporated in the recommended methodology for those situations where background is negligible and reference areas need not be chosen. Also, a median test was developed for those situations where the statistical tests indicate that radioactivity distinguishable from background is present, but where the distribution of measured concentrations suggests an average dose less than 0.15 mSv per year. Work also progressed on applying *in situ* spectrometry for assessing residual radioactivity on surfaces inside buildings. Future work will include addressing issues of biased sampling and resampling and applying unfolding routines for *in situ* spectrometry performed inside buildings to assess elevated surface area contamination.

References

Gogolak, C. V., A. M. Huffert, and G. E. Powers
"A Non-Parametric Statistical Methodology for the Design and Analysis of Final Status Decommissioning Surveys"
USNRC Report NUREG-1505 (draft) (1995)

Huffert, A. M. And K. M. Miller
"Measurement Methods for Radiological Surveys in Support of New Decommissioning Criteria"
USNRC Report NUREG-1506 (draft) (1995)

Figure 6.10 EML scientists using a collimated Ge gamma-ray spectrometry system and a GM survey meter for assessing residual surface contamination during a demonstration of newly developed final status survey methodology at a nuclear power plant.

6.21 NRC/CEC CONSEQUENCE UNCERTAINTY PROJECT

Kevin M. Miller

By invitation, EML was selected to participate, under contract, on an expert panel to support the joint United States Nuclear Regulatory Commission/Commission of European Communities Consequence Uncertainty Project. This particular panel dealt with deposited materials and related doses associated with accidental releases from commercial nuclear power plants (NRC, 1995). A formal expert elicitation process was used to provide an evaluation of the radiological consequences occurring off site using previously developed uncertainty methods. A library of distributions is being developed and quantified so that the models used in consequence codes may be evaluated for fundamental flaws in physics, data uncertainties, and stochastic uncertainties in observational data and input parameters. The panel was called together for two meetings which were held in Albuquerque, NM, and hosted by Sandia National Laboratory. Training on the elicitation of expert judgments as probability distributions was given to the panel members and case structure documentation was reviewed. Panel members then prepared a report addressing 13 questions with multiple subparts relating to such matters as radionuclide specific doses as a function of deposition process and time after deposition, location/shielding factors, relative time integrated air concentrations and population characteristics. Estimates of the median and the 5th/95th quantiles of the distributions in the results were derived. Rationale for the computational approach was provided and an oral review session was conducted with project principals. Results of the elicitation are now under review by the project team members and the lead agencies.

Reference

U.S. Nuclear Regulatory Commission/Commission of the European Communities
"Probabilistic Accident Consequence Uncertainty Analysis - Dispersion and Deposition Uncertainty Analysis"
USNRC Report NUREG/CR-6244; EUR 15855EN; SAND94-1453, Vol. 1 (1995)

6.22 THE MULTI-AGENCY RADIATION SURVEY AND SITE INVESTIGATION MANUAL (MARSSIM)

Carl V. Gogolak

The Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) is being developed collaboratively by four federal agencies having authority for control of radioactive materials: Environmental Protection Agency (EPA), Department of Defense (DoD), Department of Energy (DOE), and Nuclear Regulatory Commission (NRC). The manual will provide guidance for planning, conducting, evaluating, and documenting environmental radiological surveys for demonstrating compliance with proposed dose-based regulations for unrestricted release of sites. A multi-step process is being developed for designing and implementing radiation surveys to identify

and assess radioactive materials in structures and the environment, to support remediation of radioactive contamination, and to demonstrate compliance with existing regulations. MARSSIM's objective is to describe standardized and consistent approaches for surveys, which will provide a high degree of assurance that established dose-based release criteria, limits, guidelines, and conditions of the regulatory agencies are satisfied at all stages of the process, while at the same time encouraging an effective use of resources. The techniques, methodologies, and philosophies that form the basis of the manual are consistent with current federal limits, guidelines, and procedures. Implementation of surveys entails: (1) the use of commercially available instrumentation and equipment, and (2) survey design and evaluation of results that incorporate standard statistical approaches. EML is participating in the MARSSIM by developing the survey design and statistical analysis procedures and writing the data interpretation chapter of the manual.

6.23 INTERNATIONAL ATOMIC ENERGY AGENCY (IAEA) CONSULTANTS MEETING

Karen A. Stevenson

The IAEA held an advisory meeting (October 9-13, 1995) for the preparation of an IAEA technical document on factors relevant to the development and implementation of characterization methodologies for radioactively contaminated sites. Countries participating in this first of several such meetings included technical representatives from Australia, Canada, Denmark, the United Kingdom, and the United States. The U.S. Department of State selected Karen Stevenson of EML as the U.S. representative. At the conclusion of the meeting, a draft technical document had been produced titled "Guidance on the Characterization of Radioactively Contaminated Site." This draft document will be presented for review and revision at a second meeting currently scheduled for the spring of 1996.

7 ENVIRONMENTAL MANAGEMENT PROGRAMS

7.1 OVERVIEW

Catherine S. Klusek

In its Five-Year Plan, the DOE 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-26), has designed and is implementing an Analytical Services Program (ASP) to address these issues. Under the direction of EM-26, 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 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-26's QA programs described above, other projects 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-26 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. During 1995 several areas of analytical development utilizing liquid scintillation techniques were pursued.

Additional projects are reported which support other offices in EM. Support for the Office of Technology Development (EM-50) continued through the efforts of EML's Technical Program Manager (TPM).

EM-26 QA Program Support

7.2 EM-26 QA DOCUMENTS

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

As part of an ongoing effort in QA guidance, performance objectives and criteria for QA assessments continue to be developed. In 1995, EML presented a draft plan for assessing sampling activities, including the key elements necessary for effective programmatic control of sampling services, at the Eleventh Annual Waste Testing and Quality Assurance Symposium, American Chemical Society, in Washington, D.C.

EM-26 has developed a comprehensive assessment program for the technical and QA management activities of EM's 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. A course document was developed and has now been published (Johnson, et. al., 1995) to address concepts of the EM-26 Assessment Protocol and to complement concepts introduced in the EM-20 Auditor/Lead Auditor Training Course at DOE Headquarters. The course was designed as a three-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-26 assessment program and related protocols, and the techniques of conducting assessment interviews. The course focused mainly on the training of technical specialists for assessments using established DOE Guidance Documents DOE/0158P and DOE/0159P. Potential QA management team members were trained with established Performance Objectives and Criteria in Management Assessment Guidance Document DOE/EM-0161P.

References

Johnson, M., C.S.,Klusek, V. Pan, H. Pandya, and J. May
"EM-263 Assessment Program Training Course"
DOE Report EML-570, June (1995)

U.SDOE, EM Analytical Services Division
"Quality Assurance Guidance for Sampling in Support of EM Environmental Sampling and Analysis Activities"
Report DOE/EM-0158P, May (1994)

U.SDOE, EM Analytical Services Division
"Quality Assurance Guidance for Analytical Laboratories in Support of EM Environmental Sampling and Analysis Activities"
Report DOE/EM-0159P, May (1994)

U.SDOE, EM Analytical Services Division
"Quality Assurance Guidance for Management Assessment in Support of EM Environmental Sampling and Analysis Activities"
Report DOE/EM-0161P, May (1994)

7.3 QUALITY ASSESSMENT PROGRAM (QAP)

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

The EML Quality Assessment Program (QAP) is designed to test the quality of the environmental measurements reported by contractor laboratories to the DOE's Office of Environmental Management (EM) and the Office of Environment, Safety and Health (EH). Under QAP, real or synthetic environmental samples that have been prepared and thoroughly analyzed at EML for as many as 20 radionuclides are distributed semi-annually to the contractors and other participating laboratories. Each participant receives five samples consisting of blended contaminated soil and vegetation, and 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 1995, QAP samples were distributed on March 1 (QAP-42) and on September 1 (QAP-43). Two EML reports were issued: EML-565 (Sanderson, Pan and Greenlaw, 1995) covered the participants' results for the QAP-41 sample distribution; EML-569 (Sanderson, Greenlaw and Pan, 1995) covered the participants' results for the QAP-42 sample distribution.

During this past year, 127 laboratories received QAP samples. This was a decrease of 2 over 1994 but an increase of 18 over 1993 and an increase of 35 over 1992. One hundred and five (105) laboratories reported data for the QAP-41 sample distributions and 111 reported data for QAP-42.

The total number of analyses performed by all participants in 1995 (QAP-41 and QAP-42) was 5935, about 27% more than was reported in 1994. In general, the 1995 data reported, while satisfactory, was not as good as in 1994. For 1995, 68% of the reported results were within 20% of the EML value; 10% differed from the EML value by more than 50%. In 1994, about 75% of the reported results were within 20% of the EML value, and 8% of the results differed from the EML value by more than 50%.

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

References

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 EML-565, January (1995)

Sanderson, C. G., P. Greenlaw, and V. Pan
"Semi-Annual Report of the Department of Energy, Office of Environmental Management, Quality Assessment Program"
USDOE Report EML-569, July (1995)

7.4 GAMMA SPECTROMETRY DATA VALIDATION PROGRAM

Karin M. Decker and Colin G. Sanderson

The annual gamma spectrometry data validation program will measure the ability of various DOE laboratories to use commercially available software and in-house programs to accurately identify and quantify the nuclides in a complex spectrum. Previous work (Sanderson, 1988; Decker and Sanderson, 1992) indicated that most programs did a good job of detecting and resolving peaks when used properly, but work was needed to improve the algorithms that convert gamma-rays to nuclide concentration. The goal of this program is to assess the capability of DOE laboratories and DOE contractors to perform routine gamma spectra analysis required for EM projects, site evaluations and other DOE programs. Synthetic spectra were created using a computer code developed at the Pacific Northwest National Laboratory and converted into a variety of formats which could be read by most PC based systems. It was found that many laboratories were using stand-alone Canberra Microvax based systems which use magnetic tape for data input. The synthetic spectra were converted to this format and put on tape with help from Canberra and sent out to be tested. The spectra will now be sent out to laboratories performing EM analyses. Participants will be asked to report nuclide identities, concentrations and uncertainties, as well as Minimum Detectable Levels (MDL). The nuclear data reference used and method of calculating MDL will also be requested in order to determine possible sources of discrepancy. The laboratories will submit their results to EML for assessment and publication.

References

Sanderson, C.G.
"An Evaluation of Commercial IBM PC Software for the Analysis of Low Level Environment Gamma-Ray Spectra"
Environment International, 14, 379-384 (1988)

Decker, K.M. and C.G. Sanderson
"A Reevaluation of Commercial IBM PC Software for the Analysis of Low Level Environmental Gamma-Ray Spectra"
Applied Radiation and Isotopes, 43, 323-337 (1992)

7.5 TECHNICAL ASSISTANCE PROGRAM

Vivian Pan

The QAP participating laboratories' analytical results can be produced by different analytical methods and procedures. The DOE QAP, described in Summary No. 7.3, is a performance driven intercomparison program. That is, DOE focuses on the quality of the end analytical results independent of the analytical method rather than on results which are from prescribed analytical methods. As part of the customer support for QAP, EML provides technical assistance for participating laboratories with poor or inconsistent results. In 1995, EML conducted a technical evaluation of the Fernald Environmental Restoration Management Corporation Analytical Laboratory Services uranium analytical system in response to inconsistent performance in recent QAP evaluations. Data and document reviews were carried out to determine whether Standard Operating Procedures (SOPs) were technically accurate and appropriate for the samples and if the analytical system was biased. The evaluation concluded that the method in use was not sensitive enough for quantitative analyses at environmental levels. The agreed upon changes to the analytical method have resulted in subsequent data results with "acceptable" evaluations.

7.6 DOE MIXED ANALYTE PERFORMANCE EVALUATION PROGRAM (MAPEP)

Catherine S. Klusek, Colin G. Sanderson, Isabel M. Fisenne, Pamela Greenlaw, Anna Berne, Pamela Perry, William Rivera, Michael Johnson, William C. Rosa, Ada Kong and Yulin L.Tan

The DOE's Mixed Analyte Performance Evaluation Program (MAPEP) is administered by DOE's Radiological and Environmental Science Laboratory (RESL), ID. The program evaluates 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 the mixed analyte samples for distribution.

MAPEP will distribute samples twice a year, in June and December. Initially, the samples will alternate between a water and a solid sample. The first implementation round of MAPEP was in January 1995. The sample (MAPEP-94-W2) was a mixed radiological and inorganic water sample. EML completed the verification round analysis in August 1994. The samples were analyzed

for 5 radionuclides (^{137}Cs, ^{57}Co, ^{60}Co, ^{90}Sr and ^{239,240}Pu) and 11 priority inorganic pollutants. The results of the reference laboratories were reported in Dahlgran (1995).

During 1995, EML completed the verification rounds for MAPEP-95-S2 and MAPEP-95-W3. The MAPEP-95-S2 sample was a soil containing radionuclide and inorganic constituents. The sample was analyzed for 6 radionuclides (^{137}Cs, ^{60}Co, ^{90}Sr, ^{238}Pu, ^{239,240}Pu and ^{241}Am) and 18 inorganic elements (Al, Be, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Sb, Th, V and Zn). Inorganic data was generated using traditional atomic absorption techniques. The MAPEP-95-W3 water samples was also a mixture of radionuclides and inorganic constituents. The samples were analyzed for 10 radionuclides (^{54}Mn, ^{57}Co, ^{90}Sr, ^{137}Cs, ^{238}Pu, ^{239,240}Pu, ^{241}Am, ^{234}U, ^{235}U, and ^{238}U) and 6 RCRA elements (As, Ba, Cd, Cr, Pb and Se). Inorganic data was generated using traditional atomic absorption techniques and state-of-the-art Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) and Inductively Coupled Plasma Mass Spectroscopy (ICP-MS).

Preparations are in progress to provide support to MAPEP-96-S3 for the verification of semi-volatile organic analytes (pesticides) as well as radiological and inorganic components. A project plan has been developed to demonstrate proficiency in the testing of volatile organic constituents utilizing standard US EPA methods and measurement techniques.

References

Dahlgran, J. R.
"Mixed Analyte Performance Evaluation Program. Water Sample MAPEP-94-W2 Reference Laboratory Analytical Data," June (1995)

Rust Geotech
"Work Plan for Mixed-Analyte Performance Evaluation Soil Sample MAPEP-95-S2"
Grand Junction Projects Office Report P-GJPO-859, February (1995)

Rust Geotech
"Work Plan for Mixed-Analyte Performance Evaluation Water Sample, MAPEP-95-W3"
Grand Junction Projects Office Report P-GJPO-860, May (1995)

7.7 EXTRACTION EFFICIENCIES OF Ra, U, Th, Pu, Am,Cm AND Np IN THREE COMMERCIALLY AVAILABLE EXTRACTIVE SCINTILLATORS USING PERALS™ SPECTROMETRY

Salvatore C. Scarpitta

The extraction efficiencies (EEs) of ^{210}Pb, ^{226}Ra, ^{232}U, ^{230}Th, ^{244}Cm, ^{242}Pu, ^{243}Am and ^{237}Np into three commercially available liquid extractive scintillators have been measured under various chemical conditions. A subject paper was presented at the 41st Annual Bioassay, Analytical and Radiochemistry Conference in Boston, MA, in November 1995. The extractive scintillators were

either aliphatic amine sulfates (URAEX^R and THOREX^R) or a phosphoric acid /nitrate derivative (ALPHAEX^R). One to two milliliter aliquots of these toluene based extractive scintillators, intended for measurement in a PERALS^{TM} alpha counting system, can be mixed into a Liquid Scintillation (LS) cocktail and measured by conventional LS counting. The PERALS system rejects unwanted ß or γ signals. Using ^{232}U as a tracer, sulfuric, nitric and hydrochloric acids were tested over a range of normalities to determine their effect on the EE. The optimum EEs for URAEX and THOREX were from dilute sulfuric acid, whereas that of ALPHAEX was from nitric acid. The effect of aqueous phase pH on the EEs for each nuclide was also tested at the optimum salt (SO_4 or NO_4) and acid conditions of each extractant. Each extractive scintillator has the capability of extracting either one or several actinide elements, depending on the pH, acid normality or salt concentration of the aqueous phase. If an aqueous sample contains several alpha emitting actinides, then additional separation chemistry steps may not be required before final measurement, depending on (1) the choice of extractant, (2) chemical conditions during extraction, and (3) the alpha energy difference between co-extracting interferants and the nuclide of interest. The final choice of a yield tracer for a specific radioanalytical application would also be determined by the energy resolution of the PERALS^{TM} Spectrometer, typically 0.250 MeV.

7.8 CALIBRATION OF A LIQUID SCINTILLATION COUNTER FOR α, ß AND CERENKOV COUNTING

Salvatore C. Scarpitta and Isabel M. Fisenne

Liquid scintillation (LS) counting systems are designed to detect low-energy (e.g., ^3H, ^{14}C) to high energy ß particles (i.e., ^{90}Y, ^{106}Rh), and α particles. Samples containing mixtures of radionuclides that emit α or ß particles or conversion electrons can be detected and quantified using variations on the LS counting technique. Depending on the LS cocktail (scintillator-solvent mixture), the alpha detection efficiency is generally >95%, whereas the beta detection efficiency is dependent on energy, spectral shape and cocktail. Typically, ß particles with maximum energies (E_{max}) >0.250 MeV are detected with >90% counting efficiency.

One variation on the LS counting technique is Cerenkov counting, an adjunct to LS counting, that does **not** require a LS cocktail (Scarpitta and Fisenne, in press). Cerenkov counting may prove effective in a rapid analysis scheme for specific radionuclides (i.e., ^{210}Pb); it may provide an additional or verification methodology in the calibration of reference radionuclide solutions; and it may reduce the hazardous waste stream from the analytical operations. Cerenkov counting in aqueous samples is applicable to ß particles with endpoint energies >0.263 MeV. The Cerenkov counting efficiency is typically 40% per MeV or ß particles with endpoint energies above the Cerenkov threshold. Alpha particles are not detected in pure aqueous solutions unless an additive is used to enhance the detection counting efficiency.

Calibration data are compiled for 25 radionuclides that were individually measured in a Packard Tri-Carb 2250CA LS Analyzer (Packard Instrument Co., Downers Grove, IL) by both conventional and Cerenkov detection techniques. The relationships and regression data between the

quench indicating parameters and the LS counting efficiencies were determined using microliter amounts of tracer added to low ^{40}K borosilicate glass vials containing 15 mL of Insta-Gel XF (Packard Instrument Co., Downers Grove, IL) scintillation cocktail. Cerenkov data were obtained using various ß emitters measured in plastic vials containing a wavelength shifter solution.

In general, about 50% of the instrument background was in the 0-50 keV energy region. The average instrument background, using 20 mL plastic vials containing 10 mL of ultra-pure water, was 0.255 ± 0.018 counts per second (cps) for a 0-50 keV region of interest (ROI). The average background count rate for glass vials (0.346 ± 0.010 cps) was about 30% higher than the plastic vials. Based on replicate background measurements, the lower limit of detection (LLD) for a one-hour 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 LLD for a one-hour count, using 10 mL of a waveshifter solution, was 0.039 cps for plastic vials. The LLD, expressed as activity, ranged from 46 to 56 mBq (2.8 - 3.4 dpm) for conventional LS counting. This assumes (1) a 100% counting efficiency, (2) a 50% yield of the nuclide of interest, (3) a 1 h measurement time using low background plastic vials, and (4) a 0-50 keV ROI. The LLD may be reduced an order of magnitude if the yield recovery exceeds 90% and a lower background region (i.e., 100 - 500 keV alpha ROI) is used.

Detection efficiencies were linear over a 3 orders of magnitude range in ß activity (10 - 10,000 mBq) for both LS and Cerenkov counting. A linear relationship was observed for the Cerenkov counting efficiency (CCE) as ß energy increased from 0.300 to 2 MeV, whereas the LS efficiency was >90% for ßs with energy in excess of 0.250 MeV. A comparison of the data showed that the CCE was 20-50% less than the LS counting efficiency for ß particles with maximum energies in excess of 1 MeV.

Reference

Scarpitta, S. and I. M. Fisenne,
"Cerenkov Counting as a Complement to Liquid Scintillation Counting,"
Int. J. Appl. Rad. Isotopes, in press

Other EM Projects

7.9 COMPUTING TOTAL UNCERTAINTIES IN RADIOCHEMICAL ANALYSES

Anna Berne

When an environmental sample is analyzed for some radioactive element using radiochemical methods, the uncertainty of the calculated result has to be reported so that appropriate assessment can be made. Often, only one aliquot of the sample is taken and a single determination is made. Under those circumstances, the total uncertainty is calculated using the "propagation of errors" approach, which takes into account the uncertainties associated with the analytical procedures, as well as the counting error(s) contributed by the counting step of the method.

However, there is no agreement in the radioanalytical community as to the exact equations to use. As part of a project supported by EM-40, a systematic process has been developed to compute uncertainties in the constituent components of the analytical procedure as well as the Total Propagated Uncertainty (TPU). This was reported at the 41st Annual Bioassay, Analytical and Radiochemistry Conference in Boston, MA, in November 1995. The equations for computation have also been incorporated into a code for use in the spreadsheet application, QuattroPro™. Using the spreadsheet with appropriate inputs permitted an analysis of variations in the TPU as a function of several different variables. The relative importance of the "counting error" can be ascertained. A handbook for calculating TPUs is being prepared.

7.10 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 (TPM) again served as a technical focus group committee member for the selection of Technical Task Plans (TTP) and review of already funded programs under the Characterization, Monitoring and Sensor Technology Crosscutting Program (CMST-CP) under the EM Focus Area initiative. The meeting was held in Denver, CO, May 9-10, 1995. Principal investigators of ongoing programs gave oral progress reports to the reviewers.

8 STAFF ACTIVITIES

- **Seminar Program**
- **Awards**
- **Academic Affiliations and Activities**
- **Editorial Activities**
- **Committee and Outside Coordination Activities (National)**
- **Professional Society Activities**
- **Standards Activities**
- **Committee and Outside Coordination Activities (International)**
- **Meetings Organized**
- **Invited Lectures At Major Scientific Meetings**
- **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 on programs in other institutions through visiting scientists. The speakers and topics listed below were presented during 1995.

Mark Kritz, State University of New York at Albany
"Use of Radon to Validate Global Climate Models"
January 25

Jaak Sinnaeve, Commission of the European Communities, Belgium
"The Future of Radiation Protection Research in the European European Community"
February 8

Anton Bayer, Federal Office for Radiation Protection, Germany
"The German Federal Office for Radiation Protection and Its Tasks in the Area of Environmental Monitoring"
April 11

Robert Katz, University of Nebraska
"Theory of Heavy Ion Radio-Biology"
May 8

Jurgen Schmitz, Karlsruhe, Germany
"Radiation Exposure by Radon at Workplaces in Germany"
May 30

Gerry M. Kendall, National Radiological Protection Board, UK
"The UK Radon Programme"
June 12

Nestor Azziz, EML
"Physics Behind TLDs"
October 31

Karen A. Stevenson, EML
"Russian Field Studies"
November 7

AWARDS

Andreas C. George

Plaque presented at 1995 International Radon Symposium, Nashville, Tennessee, September 1995, for contributions to radon measurement technology

Merrill Heit

Certificate of Appreciation from National Science Foundation in recognition of service to the Interagency Arctic Research Policy Committee (IARPC)

Wayne M. Lowder (retired)

Plaque presented at 6th International Symposium on the Natural Radiation Environment, Montreal, Canada, June 1995, for contributions of research in the field of environmental radiation and radioactivity and honoring his co-founding of the series of NRE Symposia begun in 1963

ACADEMIC AFFILIATIONS AND ACTIVITIES

Isabel M. Fisenne

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

Andreas C. George

Member, Thesis Advisory Committee, Rutgers University

Carl V. Gogolak

Lecturer, Harvard University Short Course Program, School of Public Health

Adjunct Lecturer, Physics and Applied Mathematics Department, Polytechnic University

Paul E. Goldhagen

Adjunct Assistant Professor, Center for Radiological Research, Columbia University

Adam R. Hutter

Adjunct Lecturer, City University of New York

Earl O. Knutson

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

Lambros Kromidas

Adjunct Assistant Professor, College of Pharmacy and Allied Health, St. John's University

Member, Thesis Advisory Committee, Pharmacology and Allied Health, St. John's University

Robert Leifer
Member, Thesis Advisory Committee, Chemistry Department, Brooklyn College

Member, Thesis Advisory Committee, Meteorology Department, Rutgers University

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

Vincent C. Negro
Adjunct Assistant Professor, Queensborough 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 30 articles in 1995 in response to requests 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
Scientific Vice-President for Radiation Measurement, National Council on Radiation Protection and Measurements

Chair, National Council on Radiation Protection and Measurements Scientific Committee 93, "Radiation Measurements"

Chair, National Council on Radiation Protection and Measurements Scientific Committee 64-6 on "Surface Soil Contamination"

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

Member, Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) Committee

Merrill Heit

EML Representative to OHER Environmental Sciences Division (ER-74)

Member, DOE-ER Human Subjects Research Working Group

DOE Representative to Interagency Arctic Research Policy Coordinating Committee (IARPC)

Coordinator, DOE-ER Human Subjects Research Database at EML

DOE Advisor to DoD-Office of Naval Research Arctic Research Program

Member, Interagency SERDP EcoRisk Methodology Working Group

Michael J. Johnson

Member, QA Working Group, ASD Programs, DOE/EM-263, Office of Transportation, Emergency Management and Analytical Services, Office of Compliance and Program Coordination

Member, Mixed Analyte Performance Evaluation Program Working Group. DOE/EM-263, Office of Transportation, Emergency Management and Analytical Services, Office of Compliance and Program Coordination

Catherine S. Klusek

Member, QA Working Group, ASD Programs, DOE/EM-263, Office of Transportation, Emergency Management and Analytical Services, Office of Compliance and Program Coordination

Member, Mixed Analyte Performance Evaluation Program Working Group, ASD Programs, DOE/EM-263, Office of Transportation, Emergency Management and Analytical Services, Office of Compliance and Program Coordination

Earl O. Knutson

Member, Air Sampling Instruments Committee of the American Conference of Government Industrial Hygienists

Lambros Kromidas

Coordinator, DOE-ER Human Subjects Research Database at EML

Richard J. Larsen

Chair, Ground Truth Review Team, Report on Peer Review of the Conference on Disarmament, International Monitoring System

Camille G. Marinetti

Secretary, EEO/Federal Women's Program Subcommittee (New York Federal Executive Board)

Coordinator, DOE-ER Human Subjects Research Database at EML

Kevin M. Miller

Organizer, IEEE Short Course, "*In-Situ* Gamma-Ray Spectrometry for Site Characterization," 1995 Nuclear Science Symposium

Organizer, Training Course, "*In-Situ* Gamma-Ray Spectrometry" to NRC Staff and Contractors (February 1995)

Panel Member, CEC/NRC Joint Project on Uncertainty Analysis of Consequence Assessment Programs

Hemant S. Pandya

Member, QA Working Group, ASD Programs, DOE/EM-263, Office of Transportation, Emergency Management and Analytical Services, Office of Compliance and Program Coordination

Karen A. Stevenson

Member, DOE Technical Focus Group for Characterization, Monitoring and Sensor Technologies Crosscutting Program

PROFESSIONAL SOCIETY ACTIVITIES

Carl V. Gogolak

Health Physics Society, President-Elect, Radon Section

Robert Leifer

New York Academy of Sciences: Member, Advisory Committee, Meteorological Section

Vivian Pan

American Geophysical Union, Member, Committee on Public Affairs

American Institute of Physics, Member, Committee on Public Policy

Matthew J. Williamson

Health Physics Society, Secretary, Greater New York Chapter

STANDARDS ACTIVITIES

Isabel M. Fisenne

Member, Committee on Nuclear Fuel Cycle, Task Group on Environmental Methods, ASTM C26.05

Andreas C. George

Member, Radionuclides Task Group, Subcommittee on Indoor Air Quality, ASTM D22.05

Member, Working Group on Performance Specifications for Measurement of Radon in Indoor Air, ANSI N13.34

Michael J. 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 Soil and Rock, ASTM D18.01

Gladys A. Klemic

Member, Working Group, Standard for Environmental Thermoluminescent 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 Regulations 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 Low-Level Environmental Measurement Quality Assessment, ANSI 42.2

COMMITTEE AND OUTSIDE COORDINATION ACTIVITIES (INTERNATIONAL)

Harold L. Beck

United States Delegate, International Electrotechnical Commission (IEC) Technical Committee TC-45, Scientific Committee SC-45B, Member Working Groups B5 and B10

Isabel M. Fisenne

Member, Organizing Committee, International Radon Metrology Program, International Atomic Energy Agency and Commission of the European Communities

Andreas C. George

Member, Organizing Committee, International Radon Metrology Program, International Atomic Energy Agency and Commission of the European Communities

Member, International Atomic Energy Agency Coordinated Research Program for International Radon Metrology Program

Merrill Heit

United States Delegate, International Arctic Monitoring Assessment Program (AMAP), U.S. State Department

Gladys A. Klemic

Corresponding Member, European Radiation Dosimetry Group (EURADOS), Working Group 12 on "Environmental Radiation Monitoring"

Co-Chair, Environmental Monitoring Session, 11th Solid State Dosimetry Conference, Budapest, Hungary (July 10-14, 1995)

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, Organizing Committee, International Radon Metrology Program, International Atomic Energy Agency and Commission of the European Communities

Member, Fuchs Memorial Award Committee, International Aerosol Research Assembly

Philip W. Krey
Member, Principals' Committee, Global Atmospheric Watch

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 Measurements Techniques

Karen A. Stevenson
Federal Lead, Program of Joint Russian-American Field Studies (DOE/EM)

U.S. Delegate, Committee to develop International Atomic Energy Agency technical document, "Guidance on the Characterization of Radioactively Contaminated Sites"

MEETINGS ORGANIZED

Harold L. Beck
Program Committee, 31st Annual Meeting of the National Council on Radiation Protection and Measurements, April 12-13, 1995, Arlington, Virginia

Adam R. Hutter
State-of-the-Art in Measuring Soil Gas Radon and Radon Exhalation From Soil, June 12-15, 1995, EML, New York

Earl O. Knutson
State-of-the-Art in Measuring Soil Gas Radon and Radon Exhalation From Soil, June 12-15, 1995, EML, New York

INVITED LECTURES AT MAJOR SCIENTIFIC MEETINGS

Harold L. Beck
"Reconstructing Source Terms from Environmental Data" presented at the 31st Annual Meeting of the National Council on Radiation Protection and Measurements, Arlington, Virginia (April 12-13, 1995)

Andreas C. George
"Technical Keynote Address" presented at 1995 International Radon Symposium, Nashville, Tennessee (September 27-29, 1995)

Adam R. Hutter

"Radon Soil Gas Measurements" presented at the International Radon Symposium, Nashville, Tennessee (September 27-29, 1995)

Philip W. Krey

"EML's Role as the Calibration Facility for Radioactivity in the Quality Assurance Program/Science Activity Center for the Americas" presented at the Global Atmospheric Watch Workshop, Garmish-Partenkirchen, Germany (March 13-17, 1995)

"EML's Global Network and Planned Role in GAW" presented at the World Meteorological Organization-International Global Atmospheric Chemistry (WMO-IGAC) Conference on the Measurement and Assessment of Atmospheric Composition Change, Beijing, People's Republic of China (October 9-14, 1995)

VISITING SCIENTISTS

As always, 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:

Tser-Min Chang
Institute of Nuclear Energy Research, Taiwan

Amaal Ahmed Tawfik
Atomic Energy Authority, Egypt

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

Nathalie Korikian (student)
National Institute of Applied Sciences, France

EML PUBLICATIONS

1/01/95 - 12/31/95

1. Albert, B.
"Light Experimental Aircraft Aerosol Measurement Package"
USDOE Report EML-573, November, 1995

Azziz, N. : See No.(s)24

2. Beasley, T. M.
"The Inventory of Site-Derived Chlorine-36 in the Snake River Plain Aquifer, Idaho National Engineering Laboratory, Idaho"
USDOE Report EML-567, February, 1995

Beck, H. L. : See No.(s)22

3. Berne, A.
"Use of EIChroM TRU.Spec in the Determination of Americium, Plutonium and Uranium in Air Filters and Water Samples"
USDOE Report EML-575, December, 1995

Chieco, N. A. : See No.(s)22

4. Colle, R., M. P. Unterweger, P. A. Hodge, J. M. 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
"An International Intercomparison of Marine Atmospheric Radon 222 Measurements in Bermuda"
J. Geophys. Res., Vol. 100, pp. 617-638, August, 1995

5. Fisenne, I. M., A. C. George, P. M. Perry and H. W. Keller
"The April 1994 and October 1994 Radon Intercomparisons at EML"
USDOE Report EML-568, October, 1995

Fisenne, I. M. : See also No.(s)9

6. Forman, L., R. J. Larsen and R. Perkins
"Ground Truth Review"
Report on the Peer Review of the Conf. on Disarm. International Monitoring System, Expert Group Report CD/NTB/WP.224. U. S. Department of Energy, August, 1995

7. 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, February, 1995

8. George, A. C., K. W. Tu and E. O. Knutson
"Third Intercomparison of Instruments and Methods for Measuring Radon and Radon Progeny in Indoor Air"
L. Morawska, N. D. Bofinger, M. Maroni (Editors)
Indoor Air - An Integrated Approach, Elsevier Scienc Ltd., pp. 149-152, September, 1995

9. George, A. C., E. O. Knutson, K. W. Tu and I. M. Fisenne
"Intercomparison of Active, Passive and Continuous Instruments for Radon and Radon Progeny"
USDOE Report EML-577, December, 1995

George, A. C. : See also No.(s)5

10. Gogolak, C. V., A. M. Huffert and G. E. Powers
"A Nonparametric Statistical Methodology for the Design and Analysis of Final Status Decommissioning Surveys"
Draft U. S. Nuclear Regulatory Commission Report NUREG-1505, August, 1995

Goldhagen, P. : See No.(s)24, 39

Greenlaw, P. : See No.(s)34, 35

11. 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"
Science Total Environ., Vol. 161, pp. 347-361, January, 1995

Guggenheim, S. F. : See No.(s)30

Hajnal, F. : See No.(s)24, 38, 39

12. Huffert, A. M. and K. M. Miller
"Measurement Methods for Radiological Surveys in Support of New Decommissioning Criteria"
Draft U. S. Nuclear Regulatory Commission Report NUREG-1506, August, 1995

13. Hutter, A. R.
"A Method for Determining Soil Gas Rn-220 (Thoron) Concentrations"
Health Physics, Vol. 68, pp. 835-839, June, 1995

14. Hutter, A. R
"Seasonal Variations in the Radon-222 Flux to the Atmosphere"
EOS, Vol. 76, pg. 76, July, 1995

15. Hutter, A. R., R. J. Larsen, H. Maring and J. T. Merrill
"Radon-222 at Bermuda and Mauna Loa: Local and Distant Sources"
J. Radioanalytical and Nuclear Chemistry, Vol. 193, pp. 309-318, June, 1995

Hutter, A. R. : See also No.(s)4

16. Johnson, M.
"Selection and Use of Equipment for the Sampling of Liquids"
USDOE Report EML-574, November, 1995

17. Johnson, M., C. S. Klusek, V. Pan, H. Pandya and J. May
"EM-263 Assessment Program Training Course"
USDOE Report EML-570, June, 1995

Kada, J. : See No.(s)27

18. Klemic, G.
"USDOE's International Intercomparisons of Environmental Dosimeters"
Appl. Radiat. Isot., Vol. 46, pp. 515-516, August, 1995

19. Klemic, G., J. Shobe, T. Gesell and P. Shebell
"Results of the Tenth International Intercomparison of Environmental Dosemeters"
Radiation Protection Dosimetry, Vol. 58, pp. 133-142, February, 1995

Klusek, C. S. : See No.(s)17, 23

Knuth, R. H. : See No.(s)30

20. Knutson, E. O.
"Random and Systematic Errors in the Graded Screen Technique for Measuring the Diffusion Coefficient of Radon Decay Products"
J. Aerosol Sci., Vol. 23, pp. 301-310, August, 1995

21. Knutson, E. O. and P. J. Lioy
"Measurement and Presentation of Aerosol Size Distributions"
B. S. Cohen, S. V. Hering (Editors)
Air Sampling Instruments for Evaluation of Atmospheric Contaminants, 8th Edition, ACGIH, Cincinnati, Ohio, June, 1995

Knutson, E. O. : See also No.(s)7, 8, 9

22. Krey, P. W. and H. L. Beck
"EML Annual Report - Calendar Year 1994"
N. A. Chieco (Editor)
USDOE Report EML-571, August, 1995

23. Krey, P. W. and C. S. Klusek
"Soil Sampling in Complex Terrain"
J. Radioanalytical Nucl. Chem., Vol. 197, pp. 79-98, November, 1995

24. 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"
Fusion Engineering and Design, Vol. 28, pp. 534-544, September, 1995

25. 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, Air, Soil Pollution, Vol. 80, pp. 591-601, September, 1995

26. Larsen, R. J. and C. G. Sanderson
"Radionuclide Results in Air Filter Samples, 1992"
A. L. Dick, P. J. Fraser (Editors)
Commonwealth Scientific and Industrial Research Organization, Cape Grim Baseline Air Pollution Station, Baseline '92, December, 1995

27. Larsen, R. J., C. G. Sanderson and J. Kada
"EML Surface Air Sampling Program 1990-1993 Data"
USDOE Report EML-572, November, 1995

Larsen, R. J. : See also No.(s)4, 6, 15

28. Lee, H. N
"A Transport and Diffusion Model Using the Semi-Lagrangian Technique with the Spectral Method"
11th Symposium on Boundary Layers and Turbulence, American Meteorological Society, Boston, MA, pp. 176-178, April, 1995

29. Lee, H. N. and J. Feichter
"An Intercomparison of Wet Precipitation Scavenging Schemes and the Emission Rates of Radon-222 for the Simulation of Global Transport and Deposition of Lead-210"
J. Geophysical Research, Vol. 100, pp. 253-270, November, 1995

Lee, H. N. : See also No.(s)30

30. Leifer, R., R. H. Knuth, S. F. Guggenheim and H. N. Lee
"Design of the Aerosol Manifold for the Southern Great Plains Site"
Proceedings of the Fourth Atmospheric Radiation Measurement (ARM) Science Team Meeting, pp. 227-231, April, 1995

31. Maiello, M. L., J. F. Gulbin, G. de Planque and H. T. Heaton
"Ninth International (Mini) Intercomparison of Environmental Dosemeters"
Radiation Protection Dosimetry, Vol. 58, pp. 167-175, February, 1995

May, J. : See No.(s)17

32. Miller, K. M. and P. Shebell
"In Situ Gamma-Ray Spectrometry - A Tutorial for Environmental Radiation Scientists"
USDOE Report EML-557, October, 1995

Miller, K. M. : See also No.(s)12

Monetti, M.: See No.(s)25

33. Pan, V.
"Analysis of EML QAP Data from 1982-1992: Determination of Operational Criteria and Control Limits for Future PE Purposes"
USDOE Report EML-564, January, 1995

Pan, V. : See also No.(s)17, 34, 35

Pandya, H. : See No.(s)17

Perry, P. M. : See No.(s)5

Reginatto, M. : See No.(s)38, 39

34. Sanderson, C. G., P. Greenlaw and V. Pan
"Semi-Annual Report of the Department of Energy, Office of Environmental Management, Quality Assessment Program"
USDOE Report EML-565, January, 1995

35. 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 EML-569, July, 1995

Sanderson, C. G. : See also No.(s)26, 27

36. Scarpitta, S.
"A Theoretical Model for Radon-222 Adsorption on Activated Charcoal Canisters in Humid Air Based on the Polanyi Potential Theory"
Health Physics, Vol. 68, pp. 332-339, March, 1995

Shebell, P.: See No.(s)19, 24, 32

Stevenson, K. : See No.(s)40

37. Stevenson, K. A.
"Site Characterization at Low Radiation Levels"
S. Slate, R. Baker, G. Benda (Editors)
Fifth International Conference on Radioactive Waste Management and Environmental Remediation ICEM'95, Vol. 2, pp. 1597-1599, September, 1995

Tu, K. W. : See No.(s)7, 8, 9

38. Wilson, J. W., M. Reginatto, F. Hajnal and S. Y. Chun
"Calculation of Dose, Dose Equivalent, and Relative Biological Effectiveness for High Charge and Energy Ion Beams"
Health Phys., Vol. 68, pp. 532-538, April, 1995

39. 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, May, 1995

40. Wollenberg, H., C. Tsang, W. Frangos, R. Solbau, W. M. Lowder, K. Stevenson, M. Foley, E. Drozhko, G. Romanov, Y. Glagolenko, A. Posochov, Y. Yvanov, P. A. Mayak, L. Samsonova, A. Petrov, S. Ter-Saakian, N. Vasil'kova, A. Glagolev and P. Hydrospetzgeologia
"A Joint Russian-American Field Test at the Chelyabinsk-65 (Mayak) Site: Test Description and Preliminary Results"
S. Slate, R. Baker, G. Benda (Editors)
Fifth International Conference on Radioactive Waste Management and Environmental Remediation ICEM '95, Vol. 2, pp. 1381-1386, September, 1995

EML PUBLICATIONS IN PRESS

Beck , H. L.
"Reconstruction of Source Term from Environmental Measurements"
NCRP Annual Meeting Proceedings

Cavallo, A., K. Gadsby and T. A. Reddy
"Comparison of Natural and Forced Ventilation for Radon Mitigation in Houses"
Environment International

Colle, 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
"An International Marine-Atmospheric ^{222}Rn Measurement Intercomparison: Part II: Results for the Participating Laboratories"
NIST Journal of Research.

Fisenne, I. M.
"A USDOE Remediation Site Case Study"
Environment International

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

Fisenne, I. M. and H. W. Keller
"Continuous Indoor and Outdoor Measurements of ^{222}Rn in New York City"
Environment International

Fisenne, I. M., H. W. Keller and E. W. Keller
"^{210}Pb in Air of a Residence"
Health Physics

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

George, A. C., E. O. Knutson, K. W. Tu and I. M. Fisenne
"Intercomparison of Active, Passive and Continuous Instruments for Radon and Radon Progeny"
USDOE Report EML-577

Hutter, A. R.
"Spatial and Temporal Variations of Soil Gas ^{220}Rn and ^{222}Rn at Two Sites in New Jersey"
Environment International

Hutter, A. R.
"Determination of ^{222}Rn Content of the Atmosphere"
J. Lodge (Editor)
Air Sampling

Klemic, G., N. Azziz and S. Marino
"The Neutron Response of Al_2O_3:C, ^{7}LiF:Mg, Cu, P, and ^{7}LiF:Mg, Ti TLDs"
Radiation Protection Dosimetry

Knutson, E. O. and K. W. Tu
"Size Distribution of Radon Progeny Aerosol in the Working Area of a Dry Former Uranium Mine"
Environment International

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

Lagomarsino, R. J.
"An Improved Gas Chromatographic Method for the Determination of Perfluorocarbon Tracers in the Atmosphere"
Journal of Chromatographic Science

Leifer, R., L. Kromidas, R. H. Knuth and H. N. Lee
"Vertical Profile (to 20 km) of Aerosol at Alamagordo, NM, Obtained with a New Balloon-Borne Sampler"
Atmospheric Environment

Leifer, R., B. Albert, H. N. Lee, R. H. Knuth, S. F. Guggenheim and L.. Kromidas
"Aerosol Measurements at 60 m During the April 1994 Remote Cloud Study Intensive Operating Period (RSC/IOP)"
Proceedings of the 1995 Atmospheric Radiation Measurement (ARM) Science Team Meeting

Leifer, R., R. H. Knuth, S. F. Guggenheim, and B. Albert
"Aerosol Measurements at the Southern Great Plains Site: Design and Surface Installation"
Proceedings of the 1995 Atmospheric Radiation Measurement (ARM) Science Team Meeting

Monetti, M. A.
"Worldwide Deposition of 90Sr Through 1990"
USDOE Report EML-579

Pan, V. and K. A. Stevenson
"Temporal Variation Analysis of Plutonium Baseline Concentration in Surface Air from Selected Sites in the Continental United States"
J. Environmental Radioactivity

Park, T. S. and E. O. Knutson
"Evaluation of Radon Progeny Sampler Efficiency"
Radiation Protection Dosimetry

Reginatto, M., P. Shebell and K. M. Miller
"An Application of the Maximum Entropy Method for Assessments of Residual Radioactivity at Contaminated Sites"
IEEE Trans. Nucl. Sci., 1995 NSS/MIC Conference Issue

Sanderson, C. G. and P. Greenlaw
"Semi-Annual Report of the Department of Energy, Office of Environmental Management, Quality Assessment Program"
USDOE Report EML-576

Scarpitta, S. C.
"A New Beaded Carbon Molecular Sieve Sorbent for ^{222}Rn Monitoring"
Health Physics

Scarpitta, S. C. and I. M. Fisenne
"Cerenkov Counting as a Complement to Liquid Scintillation Counting"
Int. J. Appl. Rad. Isotopes

Shebell, P. and K. M. Miller
"Analysis of Eighteen Years of Environmental Radiation Monitoring Data"
Environment International

Stevenson, K. A. and V. Pan
"An Assessment of Uranium in Surface Air Within the Continental United States"
J. of Environmental Radioactivity

Tan, Y. L., A. Kong, and M. A. Monetti
"Biogenic Polycyclic Aromatic Hydrocarbons in an Alaskan Arctic Lake Sediment"
Polycyclic Aromatic Compounds

EML PRESENTATIONS

1/01/95 - 12/31/95

Beck, H. L.
"Reconstructing Source Terms from Environmental Data"
Thirty-First Annual Meeting of the NCRP, Arlington, VA, April 12, 1995

Berne, A.
"Americium Separation in Larger Soil Samples (up to 50 g)"
Eichrom Industries, Inc. Annual User's Workshop at the 41st Conference on Bioassay, Analytical, and Environmental Radiochemistry, Boston, MA, November 13, 1995

Berne, A. and H. Eng
"An Analysis of Total Propagated Uncertainties in Analytical Radioactive Measurements of Environmental Samples"
41st Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry, Boston, MA, November 14, 1995

Bogen, D. C.
"Radiochemistry Applications"
On Site Analysis, The Energy Daily and Environment Week, Crystal City, VA, March 20, 1995

Cavallo, A., K. Gadsby and T. A. Reddy
"Comparison of Natural and Forced Ventilation for Radon Mitigation in Houses"
International Symposium on the Natural Radiation Environment VI, Montreal, Canada, June 8, 1995

Fisenne, I. M.
"Continuous Indoor and Outdoor Radon-222 Measurements: A City as a Source"
International Symposium on the Natural Radiation Environment VI, Montreal, Canada, June 5, 1995

George, A. C.
"Performance and Intercomparison of Different Instruments for the Measurement of Radon and Radon Progeny"
1995 International Radon Symposium, American Association of Radon Scientists and Technologists, Nashville, TN, September 26, 1995

Gogolak, C. V.
"Methodology for Determining Whether Residual Radioactivity is Distinguishable from Background"
40th Annual Meeting of the Health Physics Society, Boston, MA, July 23, 1995

Gogolak, C. V.
"Council on Ionizing Radiation Measurements and Standards (CIRMS)"
40th Annual Meeting of the Health Physics Society, Boston, MA, July 23, 1995

Goldhagen, P.
"Radiation Safety for Future High Altitude Aircraft"
NASA Sponsored Sixth Annual Space Radiation Health Investigator's Workshop, Brookhaven National Laboratory, Upton, NY, May 3, 1995

Goldhagen, P.
"Neutron Spectrometry for Radiation Protection: Three Examples"
Fourth Annual Meeting of the Council on Ionizing Radiation Measurements and Standards (CIRMS), National Institute of Standards and Technology, Gaithersburg, MD, November 29, 1995

Goldhagen, P., F. Hajnal, M. Reginatto, J. W. Wilson and J. E. Nealy
"Radiation Safety for Future High-Altitude Commercial Aircraft"
Sixth Annual Space Radiation Health Investigators' Workshop, Brookhaven National Laboratory, Upton, NY, May 2, 1995

Hutter, A. R.
"Seasonal Variations in the Radon-222 Flux to the Atmosphere"
1995 Spring Meeting of the American Geophysical Union, Baltimore, MD, June 2, 1995

Hutter, A. R.
"Spatial and Temporal Variations of Soil Gas Radon-220 and Radon-222 at Two Sites in New Jersey"
International Symposium on the Natural Radiation Environment VI, Montreal, Canada, June 5, 1995

Hutter, A. R.
"Invited talk: Radon-222 + Radon-220 Soil Gas Measurements"
International Radon Symposium, American Association of Radon Scientists and Technologists, Nashville, TN, September 28, 1995

Johnson, M. J.
"Performance Objectives and Criteria for Field Sampling Assessments"
11th Annual Waste Testing and Quality Assurance Symposium, USEPA Offices of Solid Waste and Research and Development, Washington, D.C., July 23, 1995

Klemic, G. A.
"Invited talk: International Intercomparison of Environmental Dosimeters"
1995 Radiological Environmental Monitoring Program/Radiological Effluent Technical Specifications (REMP/RETS) Workshop, Mount Pocono, PA, June 5, 1995

Klemic, G. A., N. J. Azziz and S. Marino
"The Neutron Response of Al_2O_3:C, ^{7}LiF,Mg,Cu,P, and ^{7}LiF,Mg,Ti TLDs"
11th Solid State Dosimetry Conference in Budapest, Hungary, July 10, 1995

Klemic, G. A.
"Invited talk: "Scientific Programs of EML"
Centro De Investigaciones Energeticas, Medioambientales Y Technologicas (CIEMAT), Madrid, Spain, July 17, 1995

Klemic, G. A.
"Overview of Research and Programs at EML"
USDOE Brookhaven Area Office, Upton, NY, August 31, 1995

Klusek, C. S.
"DOE Quality Assessment Program Using Performance Evaluation Data as a Management Tool"
Fall 1995 Sample Management Office Workshop, Augusta, GA, October 25, 1995

Klusek, C. S.
"DOE QAP - An Environmental Radiological Performance Evaluation Program"
Workshop on the DOE Mixed Analyte Performance Evaluation Program (MAPEP), Augusta, GA, October 26, 1995

Knutson, E. O.
"EML's Radon Calibration Facilities and Quality Assurance Measures"
Workshop on Monitoring and Uranium Mines, Atomic Energy Control Board of Canada, Saskatoon, Saskatchewan, September 20, 1995

Krey, P. W.
"EML's Global Network and Planned Role in GAW"
WMO-IGAC Conference on the Measurement and Assessment of Atmospheric Composition Change in Beijing, People's Republic of China, October 10, 1995

Larsen, R. J.
"EML's Global Sampling Network"
1995 Monitoring Technologies Conference Sponsored by the Advanced Research Projects Agency, Chantilly, VA, May 15, 1995

Lee, H. N.
"Modeling Technique of the Semi-Lagrangian Technique Coupled with the Spectral Method for Studying Pollutant Transport, Diffusion and Chemistry"
Seminar at the Pacific Northwest Laboratory, Richland, WA, February 9, 1995

Lee, H. N.
"Global Transport of Radon-222 and Lead-210 Using Data from EML's Global Sampling Network"
Workshop on Modeling the Global Transport and Scavenging of Tracer Constituents by Clouds, Cambridge University, United Kingdom, August 3, 1995

Leifer, R. and R. H. Knuth
"Aerosol Measurements at the Southern Great Plains Site: Design and Surface Installation"
Fifth Annual Atmospheric Radiation Measurement (ARM) Program Science Team Meeting, San Diego, CA, March 20, 1995

Leifer, R. and R. H. Knuth
"Aerosol Measurements at 60 m During the April 1994 Remote Cloud Study Intensive Operating Period (RSC/IOP)"
Fifth Annual Atmospheric Radiation Measurement (ARM) Program Science Team Meeting, San Diego, CA, March 22, 1995

Miller, K. M. and P. Schebell
"*In Situ* Gamma-Ray Spectrometry"
Training Course for NRC and Contractor Staff Nuclear Regulatory Commission, Rockville, MD, February 2, 1995

Miller, K. M.
"Deposited Materials and Related Doses"
Elicitation Session, USNRC/CEC Consequence Uncertainty Project Albuquerque, NM, July 20, 1995

Miller, K. M.
"Field Measurement Methods to Support Cleanup Criteria That Approach Background Radiation Levels"
40th Annual Meeting of the Health Physics Society, Boston, MA, July 23, 1995

Miller, K. M.
"Surveys and Instrumentation"
Workshop on Development of Regulatory Guidance Implementing the Final Rule on Radiological Criteria for Decommissioning, Nuclear Regulatory Commission, Rockville, MD, September 29, 1995

Miller, K. M., P. Schebell and M. Reginatto
"*In Situ* Gamma-Ray Spectrometry for Site Characterization"
IEEE Nuclear Science Symposium, San Francisco, CA, October 25, 1995

Pan, V. and P. Greenlaw
"U. S. Department of Energy Radiological Laboratory Performance Evaluation Reports: A Management Tool for Assessing Analytical Capability"
Environmental Remediation 1995 Conference, Denver, CO, August 14, 1995

Pan, V. and W. R. Newberry
"U. S. Department of Energy Environmental Sampling and Analysis Quality Assurance Assessments"
Environmental Remediation 1995 Conference, Denver, CO, August 15, 1995

Pan, V.
"Determination of Control Limits for Analytical Performance Evaluation in USDOE's Radiological Quality Assessment Program"
11th Annual Waste Testing and Quality Assurance Symposium, USEPA Offices of Solid Waste and Research and Development, Washington, D.C., July 23, 1995

Pan, V., C. S. Klusek and W. R. Newberry
"Quality Assurance Assessments for Environmental Sampling and Analysis"
11th Annual Waste Testing and Quality Assurance Symposium, USEPA Offices of Solid Waste and Research and Development, Washington, D.C., July 23, 1995

Reginatto, M. , P. Shebell and K. M. Miller
"An Application of the Maximum Entropy Method for Assessments of Residual Radioactivity at Contaminated Sites"
1995 IEEE Nuclear Science Symposium and Medical Imaging Conference, San Francisco, CA October 26, 1995

Sanderson, C. G.
"Automatic Remote Gamma-Ray Spectrometry with Two-Way Communications"
1995 Monitoring Technologies Conference sponsored by the Advanced Research Projects Agency, Chantilly, VA, May 15, 1995

Sanderson, C. G.
"Automatic Remote Gamma-Ray Spectrometry with Two-way Communications"
ARPA Sponsored 1995 Monitoring Technologies Conference, Westfields International Conference Center, Chantilly, VA, May 18, 1995

Sanderson, C. G.
"Remote Atmospheric Gamma-Ray Spectrometry"
1995 Annual Meeting of the New England Chapter of the Health Physics Society, Westborough, MA, June 5, 1995

Sanderson, C. G.
"EML's AUTORAMP Aerosol Sampler"
ARPA Sponsored Global Atmospheric Radionuclide Detection System (GARDS) Technology Demonstration and CTBT Radionuclide Monitoring Workshop, International Data Center, Arlington, VA, November 16, 1995

Scarpitta, S. C.
"Cerenkov Counting of Environmental Radioactive Samples"
Canberra/Packard Users Group Meeting, Haines City, FL, April 4, 1995

Scarpitta, S. C. and N. Krikorian
"Extraction Efficiencies of Ra, Pb, U, Th, Pu, Am, Cm and Np by PERALS Spectrometry Using Three Commercially Available Scintillators"
41st Conference on Bioassay, Analytical, and Environmental Radiochemistry, Boston, MA, November 12, 1995

Shebell, P.
"*In Situ* Gamma-Ray Spectrometry: Past, Present, and Future"
1995 Canberra User's Group Meeting, Orlando, FL, April 1, 1995

Shebell, P.
"Analysis of Eighteen Years of Environmental Radiation Monitoring Data"
International Symposium on the Natural Radiation Environment VI, Montreal, Canada, June 1, 1995

Shebell, P.
"DOE-EML Site Characterization Studies Using In Situ Spectrometry"
1995 Russian 3rd Environmental Remediation Course, Site Characterization Group, Aiken, SC, June 5, 1995

Stevenson, K. A.
"Laboratory Update"
Nuclear Plant Program Conference, Air Force Technical Applications, Center, FL, April 4, 1995

Stevenson, K. A.
"Invited talk: Site Characterization Demonstration: MAYAK Complex"
New York Chapter of the Health Physics Society, West Point, NY, May 8, 1995

Stevenson, K. A.
"Radionuclide Contaminant Characterization Studies in the Region of Chelybinsk-65 (Ozersk)"
Vallecitos Nuclear Center, CA, August 24, 1995

Stevenson, K. A.
"Results of the U. S. Radiometric Measurements Conducted During the September 1994 Joint Russian - U. S. Field Activities"
(Head of Delegation) HYDROSPETZGEOLOGIYA State Geologic Enterprise, Moscow, Russia, September 19, 1995

Stevenson, K. A.
"Progress Report on the September 1994 Field Studies in the Territory of the MAYAK Production Facility, Russia"
Workshop of the Joint Coordinating Committee for Environmental Restoration and Waste Management (JCCEM), Washington, D.C., November 14, 1995

Tan, Y. L., A. Kong and M. A. Monetti
"Biogenic Polycyclic Aromatic Hydrocarbons in an Alaskan Arctic Lake Sediment"
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9 ABBREVIATIONS AND ACRONYMS

ACP	Atmospheric Chemistry Program
AEROCE	Atmospheric/Ocean Chemistry Experiment
AIR	atmospheric ionizing radiation
ANSI	American National Standards Institute
ANWAP	Arctic Nuclear Waste Assessment Program
AOS	Aerosol Observation System
APRF	U.S. Army Pulse Radiation Facility
ARL	Australian Radiation Laboratory
ARM	Atmospheric Radiation Measurement
ASP	Analytical Services Program
AUTORAMP	Automatic Remote Atmospheric Measurements System
BCMS	beaded carbon molecular sieve
BEIR	Committee on the Biological Effects of Ionizing Radiation
BNL	Brookhaven National Laboratory
BSM	beaded scintillation material
CART	cloud and radiation testbed
CCE	Cerenkov counting efficiency
CDIAC	Carbon Dioxide Information Analysis Center
CE	counting efficiency
CGAWBO	Chinese Global Atmospheric Watch Baseline Observatory
CMDL	Climate Monitoring and Diagnostics Laboratory
CPC	condensation particle counter
D-D	deuterium-deuterium
DLR	German Air and Space Research Institute
DNA	Defense Nuclear Agency
DoD	U.S. Department of Defense
DOE	U.S. Department of Energy

Abbreviations and Acronyms

DORT	discrete ordinates transport code
DQO	data quality objective
DREO	Defense Reserch Establishment, Ottawa
D-T	deuterium-tritium
EEs	extraction efficiencies
EH	DOE Office of Environment, Safety and Health
EM	DOE Office of Environmental Management
EML	Environmental Measurements Laboratory
EPA	U.S. Environmental Protection Agency
FMSF	Fissile Material Storage Facility
4PI	four-parallel impactor
GAW	Global Atmospheric Watch
GchM	Global Chemistry Model
GCM	General Circulation Model
GSA	graded screen array
HASP	High Altitude Sampling Program
HEU	highly-enriched uranium
HPGe	high purity germanium
HSCT	high-speed civil transport
HSD	Human Subjects Research Database
IAEA	International Atomic Energy Agency
ICP-AES	Inductively Coupled Plasma Atomic Emission Spectroscopy
ICP-MS	Inductively Coupled Plasma Mass Spectroscopy
ICPP	Idaho Chemical Processing Plant
ICRP	International Commission on Radiological Protection
ICRU	International Commission on Radiation Units and Measurements
IEEE	Institute of Electrical and Electronics Engineers
IGAC	International Global Atmospheric Chemistry

Abbreviations and Acronyms

IIIP	International Intercalibration and Intercomparison Program
IL	Isotope Laboratory
INEL	Idaho National Engineering Laboratory
IRMP	International Radon Metrology Program
JCCEM	Joint Coordinating Committee for Environmental Restoration and Waste Management
JSC	Johnson Space Center
LAHET	Los Alamos High Energy Transport Code
LAN	local area network
LANL	Los Alamos National Laboratory
LaRC	Langley Research Center
LBL	Lawrence Berkeley Laboratory
LLD	lower limit of detection
LS	liquid scintillation
MACS	marine aerosol characterization study
MAPEP	Mixed Analyte Performance Evaluation Program
MARSSIM	Multi-Agency Radiation Survey and Site Investigation Manual
MCA	multichannel analyzer
MDL	minimum detectable level
MINATOM	Russian Ministry of Atomic Energy
MLO	Mauna Loa Observatory
MOHAVE	measurement of haze and visual effects
MNS	multisphere neutron spectrometer
MOUDI	micro-orifice uniform deposit impactor
NASA	National Aeronautics and Space Administration
NCAR	National Center for Atmospheric Research
NCRP	National Council on Radiation Protection and Measurements
NIST	National Institute of Standards and Technology

Abbreviations and Acronyms

NOAA	National Oceanic and Atmospheric Administration
NRC	Nuclear Regulatory Commission
NRPB	National Radiation Protection Board
NVLAP	National Voluntary Laboratory Accreditation Program
OHER	Office of Health and Environmental Research
OPC	optical particle counter
ORNL	Oak Ridge National Laboratory
PAEC	potential alpha energy concentration
PCB	polychlorinated biphenyl
PIC	pressurized ionization chamber
PNNL	Pacific Northwest National Laboratory
PNTD	plastic nuclear track detector
PPPL	Princeton Plasma Physics Laboratory
PVB	polyvinylbenzene
PVT	polyvinyltoluene
QA	quality assurance
QAP	Quality Assurance Program
QA/SAC	Quality Assurance/Science Activity Center for the Americas
QC	quality control
RAMP	Remote Atmospheric Measurements Program
RAMS	Remote Atmospheric Measurements Systems
RARAF	Radiological Research Accelerator Facility of Columbia University
RCCC	Radiological Control Coordinating Committee
RESL	Radiological and Environmental Science Laboratory
RMC	Royal Military College of Canada
ROI	region of interest
RWMC	Radioactive Waste Management Complex
SASP	Surface Air Sampling Program

Abbreviations and Acronyms

SEM	Scanning Electron Microscopy
SGP	Southern Great Plains
TAOS	Trans-Arctic Ocean Section
TEPC	tissue-equivalent proportional counter
TFTR	Tokamak Fusion Test Reactor
TLD	thermoluminescent dosimetry
TLDs	thermoluminescent dosimeters
TPM	Technical Program Manager
TPU	total propagated uncertainty
TTP	Technical Task Plan
UOM	University of Miami
WL	working level
WMO	World Meteorological Organization
WSSRAP	Weldon Spring Site Remedial Action Project