Carlsbad Environmental Monitoring & Research Center

2002

Report

2002 Report
Carlsbad Environmental Monitoring & Research Center

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Acronyms and Abbreviations

µBq	microBecquerel
µm	micrometer
AA, AAS	atomic absorption, atomic absorption spectrometry
Ag	silver
Al	aluminum
Am	americium
AMS	accelerator mass spectrometry
ANSI	American National Standards Institute
As	arsenic
Ba	barium
Be	beryllium
Bq	Becquerel
C	centigrade
Ca	calcium
CBFO	Carlsbad Field Office
Cd	cadmium
Ce	cerium
CEMRC	Carlsbad Environmental Monitoring & Research Center
CEMRP	Carlsbad Environmental Monitoring and Research Program
CFR	Code of Federal Regulations
Ci	Curie
CIS	cooled injection system
ClO	hypochlorite
cm	centimeter
Cm	curium
Co	cobalt
COC	concentrations of concern
Cr	chromium
CRM	certified reference materials
Cs	cesium
CTC	Chemrad Tennessee Corporation
Cu	copper
DL	detection limit
DOE	U.S. Department of Energy
DOELAP	Department of Energy Laboratory Accreditation Program
DTPA	diethylene-triaminepentaacetic acid
Dy	dysprosium
EEG	Environmental Evaluation Group
EM	Environmental Monitoring
EML	Environmental Monitoring Laboratory
EPA	U.S. Environmental Protection Agency
Er	erbium
ERA	Environmental Research Associates
Eu	europium
Fe	iron
FWHM	Full Width Half Maximum
FY	fiscal year
g	gram
GC/MS	gas-chromatograph/mass spectrometer
Gd	gadolinium

Ge	germanium
GPS	global positioning satellite
HCl	hydrochloric acid
$HClO_4$	perchloric acid
HF	hydrofluoric acid
Hg	mercury
HNO_3	nitric acid
H_2O_2	hydrogen peroxide
HP	high purity
HPGe	high purity germanium
IC	ion chromatography
ICP-MS	inductively coupled plasma-mass spectrometry
ISTC	International Science and Technology Center
K	potassium
keV	kiloectron volts
km	kilometer
L	liter
La	lanthanum
LaF_3	lanthanum fluoride
LANL	Los Alamos National Laboratory
LDBC	"Lie Down and Be Counted"
LFB	Laboratory Fortified Blank
LFM	Laboratory Fortified Matrix
Li	lithium
LLNL	Lawrence Livermore National Laboratory
LRB	Laboratory Reagent Blanks
m	meter
MBL	mobile bioassay laboratory
mBq	milliBecquerel
MDC	minimum detectable concentration
MDL	minimum detection level
MeV	mega electron volts
Mg	magnesium
$MgCl_2$	magnesium chloride
MgO	magnesium oxide
min	minute
mL	milliliter
mm	millimeter
Mn	manganese
Mo	molybdenum
Na	sodium
NaCl	halite
NaOH	sodium hydroxide
Nd	neodymium
Ni	nickel
NIST	National Institute of Standards and Technology
NMSU	New Mexico State University
NORM	naturally occurring radioactive material
Np	neptunium
NRC	National Research Council
NRIP	NIST Radiochemistry Intercomparison Program
NTS	Nevada Test Site

ORNL	Oak Ridge National Laboratory
p	probability
Pb	lead
pH	scale indicating acidity or alkalinity of a substance
PM_{10}	particulate matter smaller than 10 micrometers in aerodynamic diameter
$PM_{2.5}$	particulate matter smaller than 2.5 micrometers in \ aerodynamic diameter
ppbv	parts per billion by volume
Pr	praseodymium
PRB	Program Review Board
Pu	plutonium
QA/QAP	quality assurance/quality assurance program
QAPD	quality assurance program document
QC	quality control
RPD	relative percent difference
SAB	Science Advisory Board
Sb	antimony
Sc	scandium
SD	standard deviation
Se	selenium
SE	standard error
Si	silicon
Sm	samarium
Sn	tin
Sr	strontium
$T_{1/2}$	half-life
Th	thorium
Ti	titanium
Tl	thallium
TRU	transuranic
TSP	total suspended particulates
U	uranium
UT/LS	upper troposphere and lower stratosphere
UV Vis	ultraviolet visible
V	vanadium
VOC	Volatile Organic Compound
WERC	Waste-management Education & Research Consortium
WIPP	Waste Isolation Pilot Plant
WTS	Washington TRU Solutions
XANES	X-Ray Absorption Near Edge Spectroscopy
XAS	X-Ray Absorption Spectroscopy

FOREWORD

This report was written, edited and produced collaboratively by the staff of the Carlsbad Environmental Monitoring & Research Center (CEMRC), who are hereby acknowledged for their contributions to the report and the project activities described herein. The first section is an overview of the current program activities, structure, resources and quality assurance. The second section consists of data summaries containing methods and descriptions of results of studies in the WIPP Environmental Monitoring project. Tables presenting data from the WIPP Environmental Monitoring project, and the contents of this report are available for electronic access at **http://www.cemrc.org**.

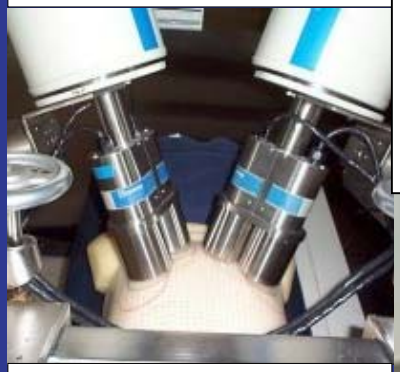
Production of this report is supported as part of the Carlsbad Environmental Monitoring and Research Center, a grant from the U. S. Department of Energy to New Mexico State University (DE-FG04-91-AL74167). The issuance of this report and other publications fulfills a major CEMRC mission in making the results of CEMRC research available for public access.

This year's cover photograph features a CEMRC radiochemist performing a step in the separation of radionuclides from environmental samples. Such radiochemical manipulations may include co-precipitation, ion exchange or extraction chromatography. Prior to the start of the separation procedure each sample is spiked with appropriate carriers and yield tracers to assist in the quantification of the radionuclides of interest. Following separation each radionuclide fraction is converted to an appropriate form for counting by gamma spectrometry, gas proportional counting, liquid scintillation counting or alpha spectrometry. The Foreword shows various equipment used by scientists at CEMRC.

Atomic Absorption Spectrometer

Inductively Coupled Plasma-Mass Spectrometer

Ion Chromatograph



Lung Counter

Nd:YAG Laser coupled with a MOPO

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X-ray Diffraction Spectrometer

Gamma Spectrometer

Oxford/Canberra Oasis Alpha Spectrometer

Dichotomous air sampler near WIPP Site

Unsaturated Flow Apparatus

OVERVIEW

Current Program Status

History and Focus

The Carlsbad Environmental Monitoring and Research Center (CEMRC) was established in 1991 with a grant from the U.S. Department of Energy (DOE). The primary goals of the CEMRC are to:

- Establish a permanent center of excellence to anticipate and respond to emerging health and environmental needs, and
- Develop and implement an independent health and environmental monitoring program in the vicinity of the DOE Waste Isolation Pilot Plant (WIPP), and make the results easily accessible to all interested parties.

The Carlsbad Environmental Monitoring & Research Center (CEMRC) is a division of the College of Engineering at New Mexico State University (NMSU). Under the terms of the grant from DOE, the design and conduct of research for environmental monitoring at the WIPP are carried out independently of the DOE, and the production and release of resulting reports do not require DOE review or approval. A brief history of the CEMRC is presented in Appendix A.

The CEMRC is operated as a research institute within NMSU, supported through grants and service contracts. The CEMRC's primary objectives are to:

- Provide for objective, independent health and environmental monitoring;
- Conduct research on environmental phenomena, with particular emphasis on natural and anthropogenic radionuclide chemistry;
- Provide advanced training and educational opportunities;
- Develop improved measurement methods, procedures and sensors; and
- Establish a health and environmental database accessible to all sectors.

Key Activities for Success

The following is a summary of progress and status for nine key enabling activities that are necessary to achieve the goal of establishing and developing the CEMRC. Activities to achieve the second goal of monitoring in the vicinity of the WIPP are presented in the following section (WIPP Environmental Monitoring Project).

1. Assemble a team of highly qualified research scientists and support staff capable of carrying out current and future projects.

At the end of 2002, the CEMRC employed 23 personnel (Table 1). No positions were vacant or in recruitment.

2. Create state-of-the-art laboratory facilities capable of supporting advanced studies in areas of scientific specialization.

In January 1997, the CEMRC was relocated to Light Hall, a new 26,000 ft^2 laboratory and office facility constructed adjacent to the NMSU-Carlsbad campus. The CEMRC's scientific activities are organized into major areas of specialization, with corresponding assignment of staff roles and responsibilities. Although some of the CEMRC's projects involve only one or two of the program areas, all of the program areas collaborate in carrying out the WIPP Environmental Monitoring project, and this type of integrative research is also applied to some newly funded projects. The five scientific program areas include (1) radiochemistry, (2) environmental chemistry, (3) informatics and modeling, (4) internal dosimetry, and (5) field programs. Detailed descriptions of each program area and associated facilities and instrumentation are presented on the CEMRC web site at http://www.cemrc.org.

3. Establish effective liaisons with leading research groups and laboratories to facilitate shared services and collaborative research.

In response to the need for expanding the CEMRC research role, the Center has developed a partnership with Los Alamos National Laboratory (LANL) to conduct actinide chemistry research for WIPP.

Program needs for external laboratory services were minimal in 2002, but some sub-

contractual agreements were maintained to provide specific specialized services or analyses (Appendix B). The NMSU Fishery and Wildlife Science Department also continued to provide support to the CEMRC through part of 2002 with the loan of a boat used in lake sampling activities. With respect to collaborative research, five of the publications and presentations by CEMRC staff during 2002 were co-authored with external colleagues, and four of the CEMRC's proposed and existing projects involve collaboration with other departments or institutions.

4. Establish an independent advisory body of scientists to provide expert guidance and consultation to CEMRC staff in the focus areas of CEMRC research.

The Scientific Advisory Board (SAB) for the CEMRC is composed of one scientific expert in each of the CEMRC's five scientific areas of specialization (Appendix C).

The Program Review Board (PRB) for the CEMRC consists of a minimum of three members selected by the NMSU College of Engineering administration (Appendix C). Members of the PRB are directors or former directors of leading environmental research centers with histories of long-term success in sponsored research.

Because of major changes in direction of CEMRC's science program, as well as a change in its leadership, the annual SAB and PRB meetings have been postponed indefinitely, starting in 2002.

5. Establish a program of administration to ensure effective operation of the CEMRC.

In July 2001, Dr. Marsha Conley, CEMRC director, retired. Dr. George Hidy acted as an interim director during the search for the new permanent director, which concluded in February 2002 when Mr. Joel Webb was appointed Director of CEMRC. Current administrative staff includes a director, an assistant to the director, a buyer specialist, a technical/facility specialist, a quality assurance manager, a word processing specialist, and an administrative secretary.

Formal tracking of CEMRC project schedules and milestones is conducted for current studies, as noted in later sections. Regularly scheduled work sessions for scientific program planning and problem solving are used to define goals and track progress. Administrative and individual program area staff also have regularly scheduled review and planning sessions. During 2002, significant accomplishments and events were reported in quarterly summaries provided to the DOE and NMSU.

6. Publish research results and create a database management system to provide access to information generated by the CEMRC.

CEMRC staff authored or co-authored 10 presentations at international, national and regional scientific meetings and 7 papers were published, are in press, or have been submitted for publication in peer-reviewed scientific journals and books during 2002 (Appendix D). A cumulative list of publications by CEMRC staff since 1996 is presented on the CEMRC web page.

The CEMRC issued a 2001 report that presented extensive data on radionuclides, non-radioactive constituents and other basic environmental parameters from the WIPP Environmental Monitoring project. This report and other CEMRC information are available via the CEMRC web site, and data tables referenced in this report are also presented on the web site at http://www.cemrc.org. Also included as part of the website are samples collected and analyzed since the most recent Center report.

7. Establish regional, national and international outreach and collaboration.

During 2002, the CEMRC hosted 6 colloquia presented by visiting scientists (Appendix E). Each colloquium was advertised locally, resulting in participation by representatives from local scientific, educational, technical and natural resource management organizations, as well as the general public. The CEMRC was involved in many other outreach activities including presentations for local civic and professional groups and exhibits for various school and

community events (Appendix F). As described in a later section, over 600 volunteers from the local community have participated in the "Lie Down and Be Counted" project. In addition, CEMRC scientists provided leadership in professional and scientific organizations and meetings (Appendix G).

8. Procure additional research grants and service contracts from external sources.

CEMRC scientists generated 10 new proposals, pre-proposals and contract modifications during 2002 (Appendix H). Important among these is the new contract modification with LANL for $52,420 for actinide chemistry research. The new funding achieved on five projects totaled over $165,000, one proposal is pending, and two proposals were not funded. A total of 13 projects (external to the CEMRC) were in progress during 2002, with a combined value over $2 million. Many of these projects and the funds received include support for external collaborators and cover multiple years of research. These projects represent a wide array of activities, and they have resulted in significant expansion and diversification of the scientific program.

9. Implement programs to offer visiting scientists training in specialized research techniques and methodologies and to involve CEMRC resources and personnel in providing educational opportunities for students nationwide.

During 2002, two undergraduate students worked in laboratory aide positions at the CEMRC; these positions provided training and basic skills development relevant to the position assignments. CEMRC staff presented 10 major presentations and special programs were provided for student groups (Appendix F).

Table 1. Listing of CEMRC Staff as of 31 December 2002

Name	Position
Arimoto, Richard	Senior Scientist-Environmental Chemistry
Brown, Becky	Assistant to the Director
Fraire, Joe	Assistant Scientist-Radiochemistry
Ganaway, David	Assistant Scientist-Field Programs
Khaing, Hnin	Assistant Analyst – Environmental Chemistry/Radiochemistry
Kirchner, Thomas	Senior Scientist-Informatics & Modeling
Lippis, Joe	Technical/Facility Specialist
McCauley, Sharyl	Quality Assurance Manager
McNutt, Damon	Programmer Analyst II
Moir, Deborah	Associate Director
Monk, James	Associate Health Physicist
Nesbit, Curtis	Associate Health Physicist
Sage, Sondra	Assistant Scientist-Environmental Chemistry
Schloesslin, Carl	Assistant Scientist-Radiochemistry
Schloesslin, Cheryl	Assistant Scientist-Environmental Chemistry
Schoep, David	Science Specialist-Internal Dosimetry
Spruiell, Roy	Programmer Analyst II
Stewart, Barry	Associate Scientist-Radiochemistry
Stroble, Carolyn	Buyer Specialist I
Walthall, Mark	Senior Scientist-Environmental Science
Webb, Joel	Director
York, Larry	Technician II-Radiochemistry
Young, Karen	Word Processing Specialist

WIPP Environmental Monitoring Project

Project Concept

As defined in the original grant, the purpose of the WIPP EM project is to establish and maintain independent environmental research and monitoring in the vicinity of the WIPP and to make the results easily accessible to all interested parties. This project was implemented during the WIPP pre-disposal phase, and is now continuing during the operational (disposal) phase. The WIPP EM project is organized and carried out independent of direct oversight by DOE, and the project does not provide data to any regulatory body to meet the compliance demonstration requirements applicable to the WIPP. Analytical results and interpretations from the WIPP EM are published by CEMRC to inform the public and particularly the environmental science community.

A detailed description of the WIPP EM concepts, sampling design and baseline studies is presented on the CEMRC web page. The following is a summary of 2001-2002 activities for each major environmental medium in the WIPP EM. It is important to note that mixed waste was first received by the WIPP on 9 September 2000. The results summarized in this report cover samples collected through October 2002.

Based on the radiological analyses of monitoring phase samples (collected since 26 March 1999) completed to date for area residents and for selected aerosols, soils, drinking water and surface water, there is no evidence of increases in radiological contaminants in the region of the WIPP that could be attributed to releases from the WIPP. In most cases, levels of radiological and non-radiological analytes measured in 2002 were within the range of baseline levels measured previously by CEMRC for the targeted analytes.

In the summer of 2001, the Carlsbad Field Office (CBFO) of DOE requested CEMRC to investigate whether the Center's direction could become more closely aligned with scientific and analytical activities foreseen by the CBFO to support the safe and efficient operation of the U.S. DOE Waste Isolation Pilot Plant (WIPP). To further develop the CEMRC program, during 2002 the Center has been working closely with the CBFO management to define research and analytical tasks that will address such needs. This redirection permits CEMRC to pursue new research avenues aggressively in partnership with (versus independent of) the DOE community in the Carlsbad region.

The tasks requested to be performed by the CEMRC during 2002 included:

- Development of a Center wide quality assurance and control program that complies with and is accountable to CBFO's Quality Assurance Program Document;
- Analytical and scientific support for the LANL Actinide Chemistry and Repository Science Program;
- Characterization of radionuclides in oil and gas from production formations in the vicinity of the WIPP;
- Identification and quantification of Gnome-derived radionuclides;
- Environmental safety and health support for WIPP operations and
- Optimization of the Center's WIPP Environmental Monitoring Project

A summary of the progress made on these tasks is also provided in this report.

Organization and Optimization of Monitoring Program

The scheduling and management of sample analyses collected in the WIPP EM project are based on (1) priorities for providing information to the public in a timely manner, (2) relative risks of human exposure to contaminants among the various media sampled, (3) needs for stringent data validation and verification prior to release and (4) time constraints resulting from sample preparation and analysis procedures.

The management plan for the WIPP EM incorporates milestones representing significant products and progress, including both routine sampling and analyses and special studies. Key performance indicators that integrate groups of milestones are

identified and reviewed annually to serve as metrics of the successful progress of the project. Completion of 2002 key performance indicators is summarized in Appendix I.

During 2002, the elements of the monitoring project were reviewed and evaluated as part of the strategic planning for CEMRC activities over the next few years. A re-definition of the scope of the monitoring program has been driven by two factors (1) the diminishing resources available for the monitoring work and (2) the increased emphasis at CEMRC on direct research and technical support of WIPP operations. The challenge that has faced CEMRC during 2002 has been to restructure and optimize the WIPP EM in order to maintain a long-term environmental monitoring program that will contribute to the public's confidence in the safe operation of the WIPP, and identify missing elements in our understanding of the WIPP environment that are not addressed by the ongoing and proposed long-term monitoring studies.

A major reduction in the resources devoted to the WIPP EM has been proposed by CEMRC through a cut back in the frequency of sampling of the various media and by reducing the number of target analytes. The justification for this reduction is based on the fact that to date, there has been no evidence for any perturbation to drinking water, soils, surface water or sediments caused by the WIPP operations. Studies of airborne particulate matter (aerosols) will continue to be a major focus of the CEMRC's monitoring efforts because, in the event that radioactive or chemical contaminants were released from WIPP, these materials could be rapidly dispersed through the atmosphere and spread throughout the environment. In response to a public survey (described in a later section), which indicated that drinking water was the environmental medium of greatest concern, CEMRC proposed annual sampling of the same drinking water sources as in prior years. While it is highly unlikely that any chemical impacts of the WIPP will be detected through analyses of other media, CEMRC considers there is value in continued monitoring of soils, surface water and sediments, and vegetation and biota at least for the remainder of the original grant. Thus, a program has been recommended in which one of the media other than aerosols and drinking water is sampled each year on a rotating basis. The decision was made to halt sampling and analysis for soils, surface water and sediments in 2002 and begin the rotational sampling of media in 2003, starting with soils.

The continuation of the WIPP EM and the optimization plans reflect the Center's commitment to ensuring that the public, workers, and the environment are protected from exposure to contaminants. Also taken into account in developing these plans was the need for the Center to move in new directions, primarily directed research and technical support of WIPP operations. It is likely that additional adjustments to the WIPP EM will be needed as the Center's capabilities continue to evolve and the other programs monitoring the WIPP also move in new directions.

Aerosols

Aerosol particle sampling is conducted at five locations, with samplers operating continuously at each location. The locations include a port inside the WIPP exhaust shaft, a site approximately 0.1 km northwest (downwind) of the WIPP exhaust shaft (On Site station), a site approximately 1 km northwest (downwind) of the WIPP (Near Field station), a site approximately 19 km southeast (upwind) of the WIPP (Cactus Flats station) (Fig. 1), and a site located in Hobbs, NM approximately 75 km northeast of the WIPP.

Continuous sampling of aerosol particles was conducted through December 2002, however, beginning in April 2002 the instruments, frequencies and locations that had been previously established in 2001 and in the baseline phase were changed to address concerns and issues and to make the sampling more efficient. Analyses of all particle samples collected through December 2002 for both radiological and non-radiological constituents were completed and are reported herein. Web site posting of results of radiological and non-radiological analyses of particle samples collected in the WIPP exhaust shaft (FAS) began in July 1999, and are

updated weekly. A summary of the 2002 FAS data is also presented herein.

The aerosol particle sampling design underwent major changes in 2002. These changes include (1) sampling for non-radiological aerosol analytes is now done through dicots only; (2) analysis for non-radiological analytes has been limited to trace elements; (3) sampling intervals for the trace elements has been lengthened to one sample per week and (4) sampling at the Hobbs aerosol monitoring station was terminated in April 2002.

Soils

During 2002, no routine soil samples were collected or analyzed.

Surface Water and Sediments

During 2002, no surface water or sediment samples were collected or analyzed.

Drinking Water

The WIPP EM studies of ground water focus on the major drinking water supplies used by communities in the WIPP region because these are often perceived by the public as a potential route for contaminants to reach humans. Five community supplies of drinking water (representing three major regional aquifers) are included in routine sampling, including Carlsbad, Loving/Malaga, Otis, Hobbs and a secondary source for Carlsbad. One private water well (representing a fourth aquifer) that is located within 16 km of the WIPP is also sampled.

During 2002, drinking water samples were collected in the spring at five of the six drinking water supplies, and results of radiological and non-radiological analyses are reported herein. The private water well was dry during the both the 2001 and 2002 sampling periods. The six drinking water supplies will continue to be sampled periodically for selected radiological and inorganic testing.

Biota

During 2002, no vegetation samples were collected or analyzed.

Human Population

The "Lie Down and Be Counted" (LDBC) project serves as a component of the WIPP EM that directly addresses the general concern about personal exposure to contaminants shared by residents who live near DOE sites. As in other aspects of the WIPP EM, *in vivo* bioassay testing was used to establish a baseline profile of internally-deposited radionuclides in a sample of local residents. The sampling design includes solicitation of volunteers from all segments of the community, with sample sizes sufficient to meet or exceed a 15% range in margin of error for comparisons between major population ethnicity and gender categories as identified in the 1990 census. Radiobioassays of the original volunteer cohort have been ongoing since July 1999. New volunteers will continue to be recruited each year, with a target of 100 new volunteers annually to establish new study cohorts and replacement of volunteer attrition.

Results of the LDBC project through 1 October 2002 are reported herein, and are updated quarterly on the CEMRC web site.

Quality Assurance

The CEMRC is subject to the policies, procedures and guidelines adopted by NMSU, as well as state and federal laws and regulations that govern the operation of the University. The management of CEMRC is committed to conducting a well-defined quality assurance program, incorporating good professional practice and focusing on the quality of its testing and calibration in research and service to sponsors. CEMRC technical programmatic areas in 2002 included: Actinide Chemistry, Environmental Chemistry, Radiochemistry, Field Programs, Informatics and Modeling and Internal Dosimetry. The development and implementation of an independent health and environmental monitoring program has been CEMRC's primary activity since establishment. As CEMRC's monitoring activities were to be conducted without direct DOE oversight, review or approval, CEMRC had not been required to comply with DOE orders or quality assurance requirements. Activities conducted at CEMRC were, however, performed in accordance with a formal system, which included a development and implementation of appropriate standards, performance assessment, quality improvement, provision of infrastructure, professional staff development, personal accountability and commitment to compliance.

Beginning in early 2002, however, a significant effort was devoted to refining CEMRC's quality system to meet applicable requirements of the US DOE Carlsbad Field Office (CBFO) Quality Assurance Program Document (QAPD, CAO-94-1012). This effort was in response to the CBFO's request for a change in CEMRC's direction to allow it to become more closely aligned with scientific and analytical activities seen by CBFO to support the safe and efficient operation of WIPP. As a result, CEMRC produced a center-wide Quality Assurance Plan (QAP) CP-QAP-004, which was subsequently submitted to and approved by DOE. In addition, one existing CEMRC document was revised and reissued, five measuring and testing equipment-related documents were written, four safety documents were revised and re-issued and ten implementation procedures, were written to address requirements of the DOE QAPD. All documents were entered into formal document control. In addition forty-five standard operating procedures from the various programmatic areas were written and issued as controlled documents, which means the controlled documents more than doubled in one year's time.

Since implementing a graded approach for conditions adverse to quality under the new QAP, one stop work, one nonconformance and thirty-two non-routine events have been documented. Most of the non-routine events occurred while implementing the QAP. All conditions adverse to quality have been addressed.

Internal surveillances were performed on the following programmatic areas: Environmental Chemistry, Field Programs, Informatics and Modeling, Internal Dosimetry and Radiochemistry. In addition, internal surveillances were performed in the Administrative, Quality Assurance areas as well as on Document Control and maintenance of Scientific Notebooks. To date, all surveillances have been closed.

During the year, external audits were performed on two of the programmatic areas at CEMRC; Internal Dosimetry (also referred to as Radiobioassay) for i*n vivo* Radiobioassay, and Environmental Chemistry for Volatile Organic Compounds (VOCs). Washington TRU Solutions (WTS) performed both of these external audits. The Internal Dosimetry Audit resulted in 4 findings and no observations. All findings were adequately addressed by CEMRC. The audit was closed in December 2002, and CEMRC was retained on the WTS Qualified Suppliers List for *in vivo* radiobioassay, lung and whole body counting. As a result of the audit on Environmental Chemistry, CEMRC was included on the WTS Qualified Suppliers List and listed as provisionally approved for VOC analysis and canister cleaning. A follow-up

audit has not yet been conducted to reflect full approval for VOC work.

The DOE CBFO performed a center-wide external audit in December 2002. The scope of that audit was to assess the status of the recently issued centerwide QAP against the requirements specified in the CBFO QAPD. This comprehensive audit included all the CEMRC laboratories and programmatic areas. The corrective action report was submitted to DOE for approval and a verification visit will be scheduled in 2003. The following sections describe and summarize the quality assurance/quality control activities of each of the laboratory programmatic areas: Radiochemistry, Environmental Chemistry, Field Programs and Internal Dosimetry.

Quality Assurance/Quality Control for Radioanalyses

Routine quality assurance/quality control activities conducted for radioanalyses include tracking and verification of analytical instrument performance, use of American Chemical Society certified reagents, use of American Society for Testing and Materials (ASTM) Type II water for reagent preparations, use of National Institute of Standards and Technology (NIST) traceable radionuclide solutions and verification testing of radionuclide concentrations for tracers not purchased directly from NIST or Analytics.

Daily (or each time the system was used) control checks were performed on all nuclear counting instrumentation. The type of instrument and methods used for performance checks were as follows. For the Tennelec LB4100 gas-flow α/β proportional counter used for the FAS program, efficiency control charting was performed using ^{239}Pu and ^{90}Sr check sources along with ensuring that α/β cross-talk was within limits. Sixty-minute background counts were recorded daily. Two blanks per week for the FAS program were counted for 20 hours and were used as a background history for calculating results. For the Protean IPC-9025 gas-flow, α/β proportional counter used for other measurements, including ^{234}Th tracer recoveries for isotopic thorium analysis, efficiency control charting was performed using ^{239}Pu and ^{99}Tc check sources along with ensuring that α/β cross-talk was within limits. Sixty-minute background counts were recorded daily.

For the Wallac Guardian 1414 liquid scintillation counter used for ^{241}Pu measurements, efficiency and centroid control charting was performed using ^{3}H and ^{14}C check sources. Fifty-minute background counts were recorded daily. Blanks counted for 12 hours were used as a background history for calculating results. Efficiency, resolution and centroid control charting were performed using ^{152}Eu check sources for the Canberra high purity germanium (HPGe) gamma detectors.

Routine background determinations were made on the HPGe detector systems by counting blank samples, and the data was used to blank correct the sample concentrations.

For the Oxford Oasis alpha spectrometer, efficiency, resolution and centroid control charting was performed using ^{148}Gd check sources on a regular basis. Before each sample count, pulser checks were performed to ensure acceptable detector resolution and centroid. Blanks counted for 5 days were used as a background history for calculating results.

During 2002, CEMRC participated in two rounds of the DOE Environmental Measurements Laboratory Quality Assurance Program (EML QAP), resulting in "acceptable" ratings for 49 individual determinations of 19 analytes in glass fiber filters, soil, vegetation and water samples (Table 2). One "warning" rating occurred for ^{40}K in soil for QAP-56 with a –15% bias. There were no occurrences of "not acceptable" ratings. Due to higher priority commitments to the development of the CEMRC quality assurance program, insufficient time was available to complete the full suite of analyses for samples from QAP-57, and thus many analytes were not reported for this set.

During 2002, CEMRC participated in the NIST Radiochemistry Intercomparison Program (NRIP) for soil, air filter and water analysis. CEMRC did not submit results for synthetic urine due to scheduling conflicts with development of the CEMRC quality assurance program. NRIP 2001 soil results were not available for the 2001 annual report and are included in this report. CEMRC was

not traceable for ^{90}Sr in any matrix with approximately –30% biases. Methods for the analysis of ^{90}Sr were still in development at the time these results were obtained. In addition, CEMRC was not traceable for ^{238}U in 2002 soil with a +13% bias and for ^{230}Th in 2002 water with a –10% bias. For all other analytes CEMRC received traceability (Table 3).

Quality Assurance/Quality Control for Environmental Chemistry Inorganic Analyses

The analytical methods employed for inorganic analyses in the environmental chemistry program at CEMRC are based, when applicable, on various standard procedures (EPA/600/4-79-020, 1983; EPA/SW-846, 1997; American Public Health Association, 1981). For some matrix/analyte combinations, appropriate external standard procedures do not exist, and for those cases, specialized procedures have been developed to meet the needs of the WIPP EM and other research projects.

Instrumentation

A DIONEX 500 ion chromatography (IC) system was used to determine the concentrations of a suite of anions, including nitrate, nitrite, sulfate, chloride, fluoride, and phosphate in water samples and aqueous extracts of aerosol samples, soils, and sediments. Configured differently, the same instrumentation was used to determine the concentrations of several cations (calcium, magnesium, sodium, ammonium and potassium). The anion analyses were performed with the use of AS11 and AS14 anion exchange columns and AG11 and AG14 guard columns, with chemical suppression and conductivity detection. The cations were determined using a CG12A guard column and a CS12A analytical column, with the same type of chemical suppression and conductivity detection.

Elemental analyses employed an atomic absorption spectrometer (AAS) with a computer-controlled Perkin-Elmer 5100PC atomic absorption unit with Zeeman background correction. Samples are introduced into the AAS by vaporization in a heated graphite furnace. Additional inorganic analyses were performed using a Perkin-Elmer Elan 6000 inductively-coupled plasma mass spectrometer (ICP-MS). The two instruments used for the elemental analyses are complementary; AAS is more sensitive than the ICP-MS for some elements, especially for the elements As and Se, but compared with the ICP-MS, the AAS has a narrower linear range, requires more operator effort for calibration and operation, and has a much lower sample throughput.

General Quality Control

Independent quality assurance samples are obtained and analyzed to verify the performance of the instrumentation and the proficiency of the analyst. Both blind samples (obtained from an outside source, with true values not known at the time of analysis) and reference samples (obtained from an outside source or prepared internally, with true values known at the time of analysis) are used to perform this function. Regular QC verifications and batch QC provide records of sample performance data. Copies of the analytical data and performance results are maintained by the environmental chemistry group.

In January 2002 the environmental chemistry laboratory participated in the WatR™ Supply Proficiency Testing Study (WS-66) sponsored by Environmental Resource Associates (ERA). All results were within the acceptance limits determined by ERA. Two ions, chloride and ortho-phosphate, results were flagged "Not Acceptable". The IC was recalibrated and the samples reanalyzed. The test sample results obtained in the reanalysis were within acceptance limits.

For all environmental chemistry analyses, QC samples are analyzed with each sample batch as an indicator of the reliability of the data produced. The types, frequencies of analysis, and limits for these QC samples have been established in a set of standard operating procedures. Extraction QC samples include Laboratory Reagent Blanks (LRB) (for aerosol and FAS samples, unused cellulose ester filters were used as LRB samples), Laboratory Fortified Blanks (LFB) (a cellulose ester

CRM, "Trace Metals on Filter Media" from High Purity Standards in Charleston, South Carolina, was used for QC of aerosol sample metals analyses), duplicates and Laboratory Fortified Matrix (LFM) samples. In cases where duplicate aliquots from the original sample were not feasible (such as aerosol filters), separate aliquots of the sample extract were analyzed for the duplicate and LFM analyses. The digestion QC parameters used for the evaluation of constituents in water, soils, and sediments were based on EPA Contract Laboratory Program (EPA 540/R-94013, 1994); and SW-846 methods (EPA/SW-846, 1997). No comparable control parameters presently exist for aerosol samples. For all constituents values were reported relative to the method detection limit as determined by the method outlined in 40 CFR 136, Appendix B.

Quality Assurance/Quality Control for Field Sampling

For the collection of most WIPP EM samples, no external standard procedures are considered completely appropriate for the objectives of the studies. In these cases, customized plans are developed and documented. After the activity is completed, the plan is revised to reflect any departures from the original plan, and documented to file. For most environmental media, the sampling plans combine selected standard procedures with specific adaptations to address scientific objectives of interest. For example, procedures for collection and preservation of samples for compliance with Safe Drinking Water Act requirements are applied to the collection of drinking water and surface water samples, but the locations of sample collection are selected on the basis of other criteria. Likewise, high-volume air samplers are operated to meet an EPA standard of 1.13 $m^3 min^{-1}$, but the frequency of filter replacement is based on optimal loading for radioanalysis.

Sampling procedures used for collection and preparation of environmental samples for the WIPP EM project are described in the individual data summaries that follow. Logbooks are maintained by technical staff in field operations to record locations and other specifics of sample collection, and data on instrument identification, performance, calibration and maintenance. Data generated from field sampling equipment are error-checked by using routine cross checks, control charts and graphical summaries. Most data collected in written form are also entered in electronic files, and electronic copies are crosschecked against the original data forms. All electronic files are backed up daily.

Calibration and maintenance of equipment and analytical instruments are carried out on predetermined schedules coinciding with manufacturer's specifications or modified to special project needs. Calibrations are either carried out by equipment vendors or by CEMRC personnel using certified calibration standards.

Quality Assurance/Quality Control for Internal Dosimetry

The *in vivo* bioassay program currently participates in, via WIPP, the Department of Energy's *In Vivo* Laboratory Accreditation Program (DOELAP) and is currently accredited as a service laboratory to perform the following direct bioassays:

- Transuransium elements via L x-ray in lungs
- ^{241}Am in lungs
- ^{234}Th in lungs
- ^{235}U in lungs
- Fission and activation products in lungs including ^{54}Mn, ^{58}Co, ^{60}Co and ^{144}Ce
- Fission and activation products in total body including ^{134}Cs and ^{137}Cs

Under DOELAP, the in vivo bioassay program is subject to the performance and quality assurance requirements specified in *Department of Energy Laboratory Accreditation Program for Radiobioassay* (DOE-STD-1112-98) and *Performance Criteria for Radiobioassay* (ANSI-N13.30). A DOELAP testing cycle was completed in 2002 that included counting phantoms representative of each of the categories listed above.

To evaluate system performance, quality control data were routinely collected throughout the year in order to verify that the lung and whole body counting system was

operating as it was at the time the system was calibrated. Quality control parameters that track both overall system performance and individual detector performance were measured. Quality control parameters tracked to evaluate individual detector performance, included:

- net peak area, peak centroid and peak resolution (FWHM) across the energy range of the spectrum,
- detector background

Quality control parameters tracked to assess overall system performance included:

- mean weighted activity of a standard source
- summed detector background

In addition, calibration verification counts were routinely performed using NIST-traceable standards and phantoms.

The Internal Dosimetry program also participated in an intercomparison study program for whole body counting administered by Oak Ridge National Laboratory (ORNL). Under this program bottle phantoms containing unknown amounts of ^{137}Cs, ^{60}Co, ^{57}Co, ^{88}Y and ^{133}Ba were sent to CEMRC, quarterly. The phantoms were counted on the lung and whole body counting system and the measured activities were reported back to ORNL and compared against the known activities. In 2002, the reported values were within –0.9% and +7.3% of the known ORNL values for all radionuclides.

Table 2. Participation in Environmental Measurements Laboratory Quality Assurance Program

Media	Radionuclide	[a]Percent Bias QAP-56	[b] Results QAP-56	Percent Bias QAP-57	Results QAP-57
Air Filter	^{241}Am	+4.2	Acceptable	[c]NR	
	^{60}Co	+1.8	Acceptable	NR	
	^{137}Cs	+4.1	Acceptable	NR	
	^{54}Mn	-0.1	Acceptable	NR	
	^{238}Pu	+6.2	Acceptable	NR	
	^{239,240}Pu	+4.1	Acceptable	NR	
	^{90}Sr	-11.8	Acceptable	NR	
	^{234}U	-0.5	Acceptable	NR	
	^{238}U	-2.0	Acceptable	NR	
Soil	^{228}Ac	-6.0	Acceptable	+1.7	Acceptable
	^{241}Am	+5.2	Acceptable	NR	
	^{212}Bi	-13.2	Acceptable	NR	
	^{214}Bi	-6.4	Acceptable	NR	
	^{137}Cs	-7.4	Acceptable	-0.4	Acceptable
	^{40}K	-15.1	Warning	-5.3	Acceptable
	^{212}Pb	-6.1	Acceptable	NR	
	^{214}Pb	-9.3	Acceptable	NR	
	^{238}Pu	+3.2	Acceptable	NR	
	^{239,240}Pu	+1.0	Acceptable	NR	
	^{90}Sr	-10.1	Acceptable	NR	
	^{234}U	-6.9	Acceptable	NR	
	^{238}U	-3.4	Acceptable	NR	
Vegetation	^{241}Am	+9.5	Acceptable	NR	
	^{244}Cm	-16.1	Acceptable	NR	
	^{60}Co	+6.0	Acceptable	NR	
	^{137}Cs	+3.0	Acceptable	+0.1	Acceptable
	^{40}K	+0.1	Acceptable	-2.2	Acceptable
	^{238}Pu	+7.2	Acceptable	NR	
	^{239,240}Pu	-0.9	Acceptable	NR	
	^{90}Sr	-4.5	Acceptable	NR	
Water	^{241}Am	+7.2	Acceptable	NR	
	^{60}Co	+1.3	Acceptable	-0.2	Acceptable
	^{134}Cs	NR		-2.8	Acceptable
	^{137}Cs	-1.5	Acceptable	+0.6	Acceptable
	Gross Alpha	-2.7	Acceptable	+11.9	Acceptable
	Gross Beta	-16.6	Acceptable	-5.4	Acceptable
	^{238}Pu	+6.0	Acceptable	NR	
	^{239,240}Pu	+3.9	Acceptable	NR	

Table continued on next page

Table 2. Participation in Environmental Measurements Laboratory Quality Assurance Program (cont.)

Media	Radionuclide	[a]Percent Bias QAP-56	[b] Results QAP-56	Percent Bias QAP-57	Results QAP-57
	^{90}Sr	-8.6	Acceptable	NR	
	^{234}U	-5.5	Acceptable	NR	
	^{238}U	-4.4	Acceptable	NR	

[a]Percent bias is calculated as the CEMRC measured value minus the EML known value, expressed as a percentage relative to the known value.
[b]Results for EML QAP "acceptable" are defined in Report EML-617, June 2002 for QAP-56 and in Report EML-618, December 2002 for QAP-57.
[c]NR = not reported, radionuclide was part of the program but not reported.

Table 3. Participation in NIST Radiochemistry Intercomparison Program

Media	Radionuclide	[a]Percent Bias	[b]Results
Soil 2001	^{241}Am	+3.1	[d]Traceable, 7%
	^{238}Pu	+1.8	Traceable, 6%
	^{239,240}Pu	+3.4	Traceable, 5%
	^{90}Sr	[c]NR	
	^{238}U	+2.9	Traceable, 16%
Air Filter 2002	^{241}Am	-0.9	Traceable, 5%
	^{238}Pu	-4.1	Traceable, 4%
	^{90}Sr	-26	Not Traceable
	^{230}Th	+3.0	Traceable, 5%
	^{238}U	-0.9	Traceable, 5%
Soil 2002	^{241}Am	+0.5	Traceable, 5%
	^{238}Pu	-3.0	Traceable, 4%
	^{90}Sr	-30	Not Traceable
	^{230}Th	-1.5	Traceable, 12%
	^{238}U	+13	Not Traceable
Water 2002	^{241}Am	-2.0	Traceable, 5%
	^{238}Pu	-1.7	Traceable, 7%
	^{90}Sr	-30	Not Traceable
	^{230}Th	-10	Not Traceable
	^{238}U	+1.1	Traceable, 3%

[a]Percent bias is calculated as the CEMRC measured value minus the NIST known value, expressed as a percentage relative to the known value.
[b]Results and traceability limit (expressed in percent) for NIST Traceablity are defined under ANSI 42.22.
[c]NR = not reported, radionuclide was part of the program but not reported.
[d]ANSI N42.22 defines the acceptance criteria for traceability to NIST for performance testing as:

$$|V_C - V_N| < \overbrace{3 \times \sqrt{\sigma_C{}^2 + \sigma_N{}^2}}^{\textit{Traceability Limit}}$$

V_C = CEMRC Value.
V_N = NIST Value.
σ_C = 1 sigma total uncertainty of V_C.
σ_N= 1 sigma total uncertainty of V_N

WIPP ENVIRONMENTAL MONITORING DATA SUMMARIES

Aerosol Studies for the WIPP-EM

History and Evolution of the Aerosol Program

The CEMRC aerosol particle (henceforth simply "aerosol") studies mainly focus on man-made and naturally-occurring radionuclides (including those known or expected to occur in the wastes to be deposited at the WIPP), but the studies also have included some investigations of selected non-radioactive, inorganic constituents. Results of the aerosol studies have been summarized in each of the Center's Annual Reports starting in 1997, and in a paper based on the WIPP-EM aerosol research which was entitled "*^{239,240}Pu and Inorganic Substances in Aerosols from the Vicinity of the Waste Isolation Pilot Plant: The Importance of Resuspension*," (Arimoto, et al., 2002).

Since the inception of the WIPP-EM program in 1996, samples for the radionuclide studies have been collected using high-volume samplers ("hivols," flow rate ~1.13 m^3 min^{-1}). The first three aerosol stations to be established (On Site, Near Field and Cactus Flats) each supported a hivol sampler collecting total suspended particulate (TSP) matter, and the Near Field and Cactus Flats stations also supported a second hivol sampler collecting particulate matter less than 10 μm aerodynamic equivalent diameter (PM_{10}, see Table 4). Hivol TSP samples were collected at a fourth station (Hobbs) from 26 September 2000 to 6 April 2002. The sampling design for the aerosol/radionuclide studies was to collect the maximum amount of particulate material as reasonably practical, and to do this individual samples typically have been collected over intervals ranging from three to six weeks depending on the aerosol loadings.

Sampling for the WIPP-EM aerosol studies has undergone a series of changes as the program has evolved and as new issues and concerns have been identified and addressed. Table 4 shows the various types of aerosol samplers that were in use at the ambient aerosol stations at the beginning of FY 2002. Up until the end of March 2002, low-volume samplers ("lovols," 10 L min^{-1}) and Graseby-Anderson dichotomous samplers (dichots) were used for collection of aerosols for non-radioactive, inorganic constituents, specifically trace elements and selected ions. The analytical methods for inorganic constituents do not require as large amounts of sample as do the radionuclide analyses, and therefore until the aerosol sampling program underwent major changes, two 2-day samples and one 3-day sample were collected each week for the non-radioactive analytes.

The WIPP-EM underwent major restructuring in FY 2002, and sampling for the non-radiological aerosol analytes is now done using dichots exclusively. Furthermore, the non-radiological analytes have been limited to trace elements, and the sampling intervals for the trace elements have been lengthened to one sample per week. More detailed information on the changes in the aerosol studies is presented in the New Directions section below.

In addition to the ambient studies, aerosols also have been and continue to be collected using a fixed air sampler (FAS) in the WIPP exhaust shaft. The results of the FAS studies are presented separately in this annual report.

In the interest of better interpreting the ambient aerosol and FAS results, the activities of ^{210}Pb and ^{7}Be were determined for a subset of the high-volume samples. ^{210}Pb and ^{7}Be are two naturally occurring radionuclides (or naturally occurring radioactive materials, NORMS) that have been used as tracers of air mass history (e.g., Graustein and Turekian, 1996). ^{210}Pb (half life, $\tau_{1/2}$ = 22 years) is produced in the atmosphere via the radioactive decay of the noble gas ^{222}Rn, which in turn is a decay product of primordial ^{238}U. ^{222}Rn evades from soils and decays in the atmosphere through several short-lived daughters eventually forming ^{210}Pb, which rapidly attaches to fine aerosol particles. In contrast, ^{7}Be is produced in the stratosphere and upper troposphere as a result of spallation reactions involving cosmic rays. On an annual basis ~75% of the ^{7}Be in surface air in the mid-latitudes is from the upper troposphere with

the remainder being stratospheric in origin (Dutkiewicz and Husain, 1985).

As a consequence of their unique sources, the ratio of ^{7}Be to ^{210}Pb is higher in air parcels with stronger influences from the upper troposphere and lower stratosphere (UT/LS), that is, the ratio can be viewed as an index of vertical mixing. In this context, model calculations (Koch et al., 1996) indicate that the ^{7}Be/^{210}Pb ratio over the continents should reach a maximum in summer as a result of dry convection. The ^{7}Be and ^{210}Pb data can therefore be used to assess possible contributions of residual materials from stratospheric nuclear weapons tests to the actinide activities at our surface sites. Although the importance of the UT/LS source for contemporary aerosol actinides has been discounted on theoretical grounds (Lee et al., 1998), no data have been collected which actually verify a lack of effect of stratospheric mixing on the contemporary activities of the actinides.

In the next two sections of this chapter, we briefly review the methods used for the aerosol studies and then summarize some recent results, highlighting the continuing studies of radionuclides and trace elements, including our first reported data for ^{241}Am, and also introducing the new data for ^{7}Be and ^{210}Pb. The chapter concludes with a summary of the new directions for the aerosol sampling program.

Methods

Detailed information regarding the sampling design for the WIPP EM ambient aerosol studies has been presented in the CEMRC reports for 1998 to 2001. Briefly, several different types of aerosol particle samples were collected at three sampling stations, On Site, Near Field and Cactus Flats (Fig. 1). For the radionuclide studies, high-volume samples were collected on 20 x 25 cm Gelman A/E™ glass fiber filters. Gravimetric measurements of the glass fiber filters were made to determine the mass of aerosol material that accumulated over the sampling interval.

The high-volume samples were analyzed for selected radionuclides, including ^{238}Pu, ^{239,240}Pu and ^{241}Am following 4 hr of heating in a muffle furnace at 500° C, which drives off organics; dissolution of the material on the filters using strong acids (HF, HCl and $HClO_4$); and multiple precipitation, co-precipitation, and ion-exchange and/or extraction chromatography steps. The nuclides of interest were precipitated with LaF_3, deposited onto filters, mounted on planchettes, and counted using an Oxford Oasis alpha spectroscopy system. The radionuclide data are reported in the following two ways. First, the *activity concentration* is calculated as the nuclide's activity per unit volume of air sampled (Bq m^{-3}). Second, *activity density* is calculated as the nuclide's activity per unit aerosol mass collected (Bq g^{-1}). Minimum detectable activities for the actinides in the high-volume aerosol samples are presented in Table 5.

^{210}Pb, and ^{7}Be activities were determined by gamma counting of the PM_{10} high-volume filters. These nuclides are known to be concentrated on submicrometer aerosols (Winkler et al., 1998), and thus the PM_{10} samples should reflect their activities in the atmosphere as well as TSP samples would. ^{210}Pb was determined using a γ-ray spectrometer consisting of a low-background shield and two dual crystal [NaI(Tl) and CsI(Tl)] scintillation detectors. A count time of 30 minutes and the 46.5 keV γ-ray was used for quantitation of ^{210}Pb. ^{7}Be also was determined by γ-ray spectrometry (477 keV γ-ray) with a count time of 30 minutes; however, in this case the counting was done using an array of 4 coaxial hyper-pure germanium detectors, each independently coupled to a 15-liter, low-background, stainless steel cryostat. These detectors are essentially cylinders of high purity (HP) Ge with an N contact on the outer surface, and a P contact on the surface of an interior axial well. The N and P contacts consist of diffused lithium and implanted boron, respectively. The HP Ge has a net impurity level of 10^{10} atoms cm^{-3}. This trace level of impurities results in depletion of the entire detector volume with moderate bias, resulting in maximum efficiency of charge collection.

For the trace element (TE) studies, the dichot and lovol aerosol samples were prepared for analyses using a mixture of

strong acids (HCl, HNO_3, and HF) and H_2O_2, and heat and high pressure in a microwave digestion system. The concentrations of major and trace elements were determined by atomic absorption spectrometry (AAS) and inductively-coupled plasma mass spectrometry (ICP-MS). The magnitudes of the uncertainty components, except flow volumes, vary among the inorganic analytes, and representative values for each range from < 5% to 15%, resulting in combined uncertainties of 15% to 20%. Representative detection limits are as follows: Al = 1×10^{2} ng m^{-3}, Th = 1×10^{-3} ng m^{-3} and U = 4×10^{-3} ng m^{-3}.

Results and Discussion

Brief Review of Prior Results

One of the most interesting and important findings from the prior WIPP-EM aerosol studies was that ^{239,240}Pu in aerosols from all stations exhibited seasonal patterns that mirrored those of aluminum, an element often used as an indicator of mineral dust (Arimoto et al., 2002). Results presented in that paper showed no evidence for an impact of the WIPP on ^{239,240}Pu activity concentrations, and a combination of chemical/radiochemical and meteorological data indicated that it was the resuspension of contaminated soil particles that was the most likely source for ^{239,240}Pu in the aerosols. Most of the samples for that study were collected before the WIPP became operational, and therefore the radionuclides were assumed to be from the global fallout produced by atmospheric weapons testing.

Summary Statistics and Seasonality

Summary statistics for the actinide activities in high volume aerosol samples are presented in Table 6. Inspection of the table shows that the highest activities of all nuclides quantified, both in terms of activity concentrations and activity densities, occurred not at the On Site station, which is where one would expect any influences from the WIPP to be strongest, but rather at Cactus Flats, which is the reference station, (in the case of the ^{238}Pu activity concentrations, for which there are only a few data points, the arithmetic means for Cactus Flats and On Site were the same). The most parsimonious explanation for these results is that the actinide concentrations have not been measurably influenced by activities at the WIPP, i.e., there has been no discernable impact of the WIPP on the activities of any of the actinides quantified.

The ^{241}Pu activities were below the minimum detectable values for all of the 215 samples analyzed as of April 2003. ^{238}Pu was infrequently detected, with activity concentrations above minimum detectable levels in only six of the 215 samples. ^{239,240}Pu was above detection limits in 214 of the 215 samples, and as in prior years, the ^{239,240}Pu activity concentrations showed a strong annual cycle (Fig. 2), with increases in activities coinciding with peaks in Al (dust) concentrations. In the earlier studies, sporadic peaks in ^{239,240}Pu activities occurred in late fall/early winter, especially in 1999, but no such peaks were observed in the data for 2000, 2001 or 2002.

The peak ^{239,240}Pu activities generally occur in the March to June timeframe, which is when strong and gusty winds in the area frequently give rise to blowing dust. Plots of both the fine and coarse dichot data for Al (Fig. 3) clearly show the same types of springtime peaks in dust that were seen previously when sampling was done using lovols. The Al concentrations were similar at all stations, and this is an indication that there have been no significant effects on alumino-silicate dust loads caused by activities at the WIPP. Furthermore, the fact that the dichots capture the seasonality of the atmospheric dust cycle ensures continuity in the database and provides a basis for comparison for future trends. Time-series plots of various elements (Fe, Si, rare earths, U, and Th: not shown) indicate that the seasonality in their concentrations follow the same seasonal cycle as Al.

Methods for determining the activity of ^{241}Am were developed by the CEMRC radiochemistry group over the past few years, and the first ambient aerosol data for this nuclide are presented here. The activity concentrations of ^{241}Am in the high-volume samples closely tracked those of ^{239,240}Pu as shown in Fig. 2. Most notably, strong

springtime peaks in ^{241}Am activity concentrations were evident in the samples from 2001 and 2002.

In contrast to the actinide and mineral dust data, the aerosol mass loadings at On Site were the highest of the three stations with comparable data sets (Table 6, note that the Hobbs data set is much smaller and therefore is not directly comparable with the results from the other stations, but in the cases where data from all stations is available, the mass loadings at Hobbs often were the highest). A timeseries plot (Fig. 4) shows that the aerosol mass loadings at all stations tend to track one another remarkably well, but that during several extended periods, most noticeably January 1999 to July 2000 and July 2001 to January 2002, the mass loadings at On Site were consistently higher than at the other sites.

As a consequence of the similar ^{239,240}Pu activity concentrations at all stations and the higher mass loadings at On Site, the activity densities at On Site tended to be lower than at Cactus Flats or Near Field (Table 6 and Fig. 5; again, the data from Hobbs are not as extensive and no comparisons have been made relative to that site). The combination of ^{239,240}Pu and gravimetrics data thus suggest that activities at the WIPP may in fact generate detectable levels of aerosol particles, but those particles actually contain less ^{239,240}Pu than typical ambient aerosols. One might speculate that these particles could be construction dusts or salt from the underground operations.

The ratio in the activity of the two NORM air mass tracers also showed strong seasonality, with a peak in the ^{7}Be/^{210}Pb ratio occurring in the May/June timeframe (Fig. 6), demonstrating that this index of vertical mixing is indeed highest when convection is strongest. The observed activity ratio of the two nuclides (arithmetic mean for twenty-three samples = 9.97) is consistent with the model of Koch (Koch et al., 1996), which predicts that the ^{7}Be/^{210}Pb ratio over the southwestern USA should be between 5 and 10. These authors also showed that the ^{7}Be/^{210}Pb ratio should increase with the elevation of the sampling site due to the dependence of ^{7}Be production with altitude. This is relevant for the Carlsbad data because the sites used for WIPP-EM sampling are at elevations of ~3000 ft or more above sea level.

Relationships among Analytes

A more direct way of visualizing relationships among analytes than by comparing times-series plots is through the construction and examination of X-Y scatterplots. The relevance and interpretive value of mineral dust data for studies of aerosol radionuclides can be illustrated through scatterplots of U vs. Al for both fine and coarse particles (Fig. 7a-b). These figures show that the concentrations of U are highly correlated with those of Al in both particle size fractions (r^2 = 0.92 and 0.85, for the coarse and fine fractions, respectively). Furthermore, these plots show that there are no apparent differences between On Site and Near Field or Cactus Flats, indicating no impact of the WIPP on U. The U to Al mass ratios, determined through regression analysis were 4.8×10^{-5} for the coarse samples and 2.5×10^{-5} fine samples from all stations combined. These values are similar to the U/Al ratio (3.5×10^{-5}) in the Earth's upper continental crust (Taylor and McLennan, 1995). The combination of the U and Al data thus support the existence of a dominant, natural, mineralogical source for U. Plots of Th vs. Al (not shown), similarly demonstrate that the Th concentrations also are highly correlated with Al, and therefore the activity of this naturally occurring alpha-emitter also is largely determined by the amount of windblown mineral dust in the atmosphere.

^{238}Pu was detected relatively infrequently; however, in those cases where ^{238}Pu was detected, its activity tended to increase with ^{239,240}Pu (Fig. 8a).

^{241}Am and ^{239,240}Pu activities were highly correlated, and the activities of these two nuclides were similar at all stations (Fig. 8b). The intercept for the ^{241}Am vs. ^{239,240}Pu regression for the hivol data from all stations combined was 9.7×10^{-10}; the slope was 0.31; and the coefficient of determination, r^2, was 0.78. Converting the activity ratio into a functional form (allowing for errors in both variables) through the application of orthogonal regression {i.e., dividing the slope from the simple least-squares regression by

the correlation coefficient (r)} results in a $^{241}Am/^{239,240}Pu$ activity ratio of 0.35. This is similar to what one would predict from the radioactive decay of ^{241}Pu and the subsequent ingrowth of ^{241}Am, implying there has been no fractionation of the nuclides.

As a point of comparison for the WIPP EM aerosol data, (Vintró et al., 1999) considered the $^{241}Am/^{239,240}Pu$ ratio in soils as part of their study of nuclide fluxes in waters of the western Mediterranean and Strait of Gibraltar. These authors noted that in 1974 the inventory of ^{241}Am in soils was 22% of that of $^{239,240}Pu$, and they further observed that if no fractionation of the nuclides were to occur, the percentage of ^{241}Am relative to $^{239,240}Pu$ would theoretically increase to 42% in 2037 simply due to the physics of ingrowth and radioactive decay. These authors also noted that the $^{241}Am/^{239,240}Pu$ ratio for mid-latitude soils in the late 1980s was 0.37 and that in 2000 the $^{241}Am/^{239,240}Pu$ value for soils would reach ~0.40. As noted above, these latter values are quite close to the ratio of 0.35 observed for the WIPP EM aerosols.

The Al mass loadings and the $^{239,240}Pu$ activity concentrations were positively correlated even though the samples were not precisely matched in time (Fig. 9). As noted above, the radionuclide samples were collected over much longer intervals than the elemental samples were. This was because the sampling design called for alternating trace element and major ion samples at least for part of study, and as a result, the weighted-average Al data for many of the samples cover only about half of each interval covered by a hivol radionuclide sample. Interestingly, the tightest correlation between Al and $^{239,240}Pu$ was found for the On Site samples where the coefficient of variation, r^2, was 0.61; this can be interpreted to mean that more than half of the variance in the $^{239,240}Pu$ activity concentrations at the On Site station can be explained by the loadings of mineral dust.

The similarity in $^{241}Am/^{239,240}Pu$ ratios for aerosols and soils combined with the matching seasonal cycles of the actinides and Al further supports the hypothesis that previously-contaminated resuspended soils are the dominant source for these nuclides in aerosol particles in the Carlsbad/WIPP area. Thus in the same way that correlations between U/Al and Th/Al can be used to infer a geological origin for U and Th, the elemental analyses also provide information useful for identifying the underlying cause for the seasonality in the actinide data.

Plots for the $^{7}Be/^{210}Pb$ ratio vs. $^{239,240}Pu$ (not shown) indicate that the observed seasonal patterns in the air mass tracers of vertical mixing have little or no explanatory power for aerosol $^{239,240}Pu$. The correlations for these two variables vs. $^{239,240}Pu$ were not significant (i.e., $p > 0.05$) for either the Near Field or the Cactus Flats sites.

New Directions for the Aerosol Program

The scope of the WIPP-EM was reviewed as one of the task-oriented projects undertaken by CEMRC in 2002. The desire to optimize the WIPP-EM was driven by two factors (1) the diminishing resources available for the monitoring work and (2) the increased emphasis at CEMRC on direct research and technical support of WIPP operations. There were two ways in which the scope of aerosol studies was scaled back; these were to (1) reduce the number of samples collected and (2) reduce the number of target analytes. The number of aerosol samples was reduced by eliminating the Hobbs sampling station and also by reducing the number of samples collected at the other stations.

Operational aspects of the streamlined aerosol component of the WIPP EM are summarized in Table 7. For the redesigned studies, the same types of collectors, i.e., a high volume TSP and a dichotomous sampler, are used at three stations On Site, Near Field, and Cactus Flats. As noted, the high-volume TSP samplers are for mass and radionuclide determinations while the dichotomous samplers are for trace elements. The decision to use high-volume TSP samplers for the radionuclide studies rather than PM_{10} samplers (which also were used previously) was made in light of the overall goal of the monitoring program, i.e., to detect any potential releases from WIPP. The decision to use TSP rather than PM_{10} samplers for the radionuclide studies reverts back to the overall objective of the WIPP-EM, which is to evaluate any

possible impacts of the WIPP. In particular, this decision was made because it could be argued that the PM_{10} samplers would not capture any releases of the largest aerosol particles as effectively as the TSP samplers.

The decision to switch from the low-volume aerosol samplers ("lovols") to the dichotomous samplers ("dichots") for the non-radioactive constituents was based on the following technical and practical reasons:

(1) The dichot sampler is commercially available, and it is approved by the U.S. EPA whereas the lo-volume samplers are built based on a proprietary design developed by Harvard University.

(2) The dichots cut the aerosol population in a single air stream into two fractions, a "fine" fraction, which is less than 2.5 μm in an aerodynamic diameter and a "coarse" fraction 2.5 to 10 μm. This feature means that in the dichot samplers, the entire PM_{10} fraction is split between two filters. In comparison, the individual PM_{10} and $PM_{2.5}$ lovol units sample separate air streams, and their cutoff characteristics are such that additivity is not assured (for example, the $PM_{2.5}$ cannot be subtracted from the PM_{10} to yield unambiguous information on the $PM_{2.5}$ to PM_{10} fraction).

(3) Flow rates of the dichots are higher than the lovols, thus providing more material for analysis.

(4) The dichot sampler uses smaller filters (37 mm vs. 47 mm diameter). Thus lower blanks would be expected.

As a further reduction in the scope of the WIPP EM, the number of trace element analytes for the aerosol studies was reduced to those elements listed as components of the mixed waste (Ag, As, Ba, Be, Cd, Cr, Pb, Hg, and Se) plus three indicator elements (Mg, Na and Al). Of the indicators, information on Mg may be useful owing to the use of MgO as a backfill in the repository, Na is an indicator of halite (NaCl), which is a major constituent of the salt caverns, and Al is an indicator of mineral matter as discussed above.

As a result of an internal program evaluation, the decision was made to eliminate the ion chromatographic analysis. This decision was based on the limited applications of the major ion data to issues concerning the WIPP. While these data are undoubtedly useful for understanding the sources for aerosols and to some extent the dynamics of the major biogeochemical cycles of sulfur and nitrogen, there is no evidence that the major ions (other than possibly Na^+), are either impacted by the WIPP or that their concentrations co-vary with any substances of concern, especially the radionuclides. Initially, it was expected that there might be correlations between the man-made radionuclides and nitrate and sulfate due to anthropogenic perturbations of the nitrogen and sulfur cycles, but this has proven not to be the case.

Summary Statements

The results presented here demonstrate that elemental data collected for the CEMRC monitoring program provide information directly relevant to understanding the seasonal cycles in aerosol-associated radionuclides from the WIPP environs. More specifically, the Al data can be used to demonstrate that the activities of both naturally occurring (U and Th) and man-made (^{241}Am and $^{239,240}Pu$) radionuclides in the atmosphere track those of mineral dust quite closely. The WIPP-EM studies have shown that more than half of the variability in the $^{239,240}Pu$ activities at the On Site station (and significant fractions of the variance at Cactus Flats and Near Field) can be explained by the seasonal cycles of blowing dust. For U and Th, the predictive value of Al is even higher: ~80% to over 90% of the variance in these nuclides can be explained by their relationship to dust.

The implications of these findings are significant for future monitoring of the WIPP. Dust concentrations vary strongly over decadal timescales, largely in response to cycles of drought in the dust source regions (Prospero and Nees, 1997). Any changes in atmospheric dust loads driven by changes in climate would presumably be reflected in the actinide concentrations observed at the WIPP. Without supporting elemental data, the causes for any increases in actinide activity concentrations could easily be misinterpreted.

Table 4. Aerosol Sampling Status for WIPP EM (March 2002)

Site	[a]Sampler	Analysis	Frequency	Comments
Station A (Exhaust Shaft)	PM_{10}-Shrouded Probe	Mass, Gross Alpha and Beta Activities, Trace Elements, Gamma Emitters, Actinides	Daily	Continuous
On Site	TSP-HI VOL	Mass & Radionuclides	[b]Variable	Continuous
	LTSP	Trace Elements (TE) & Ions (I)	4,3 Day Cycle	Cycle TE/I/I/TE
	DICHOT	Trace Elements (TE) & Ions (I)	4,3 Day Cycle	Cycle TE/I/I/TE
Near Field	TSP-HI VOL	Mass & Radionuclides	Variable	Continuous
	PM_{10}-HI VOL	Mass & Radionuclides	Variable	Continuous
	LTSP	Trace Elements (TE) & Ions (I)	4,3 Day Cycle	Cycle TE/I/I/TE
	DICHOT	Trace Elements (TE) & Ions (I)	4,3 Day Cycle	Cycle TE/I/I/TE
Cactus Flats	TSP-HI VOL	Mass & Radionuclides	Variable	Continuous
	PM_{10}-HI VOL	Mass & Radionuclides	Variable	Continuous
	LTSP	Trace Elements (TE) & Ions (I)	4,3 Day Cycle	Cycle TE/I/I/TE
	DICHOT	Trace Elements (TE) & Ions (I)	4,3 Day Cycle	Cycle TE/I/I/TE
Hobbs	TSP-HI VOL	Mass & Radionuclides	Variable	Continuous

[a] Sampler types are as follows: PM10-Shrouded Probe = particles greater than 10 μm diameter (50% cut-size), TSP-HI VOL = high volume total suspended particles, PM10-HI VOL = particles greater than 10 μm diameter (50% cut-size), LTSP = low volume total suspended particles and DICHOT = dichotomous sampler.

[b] Samples are changed when the flow drops to 90% of original for the 2-stage pumps.

Table 5. Minimum Detectable Activities (MDAs) for Actinides in High-Volume Aerosol Samples

MDA	Units	^{241}Am	^{238}Pu	$^{239,240}Pu$	^{241}Pu
Activity Concentration	Bq m^{-3}	3.1E-09	3.9E-09	2.6E-09	1.2E-06
Activity Density	Bq g^{-1}	1.1E-04	1.3E-04	8.5E-05	4.2E-02

Table 6. Summary Statistics for Aerosol Mass Loadings and Actinide Activities in High-Volume Aerosol Samples

Station		Cactus Flats		Hobbs	Near Field		On Site
Type of Sample		PM_{10}	TSP	TSP	PM_{10}	TSP	TSP
Number of Samples		32	50	11	28	49	45
Aerosol Mass, micrograms per cubic meter	[a]N	32	50	11	28	49	45
	Mean	0.83	1.45	2.09	0.77	1.35	1.66
	StdDev	0.27	0.60	0.89	0.25	0.53	0.59
^{241}Am Activity Concentration, Bq m^{-3}	N	NA	14	6	NA	11	13
	Mean	NA	7.0E-09	4.5E-09	NA	4.4E-09	5.5E-09
	StdDev	NA	3.9E-09	1.1E-09	NA	2.3E-09	2.4E-09
^{241}Am Activity Density, Bq g^{-1}	N	NA	14	7	NA	11	14
	Mean	NA	1.9E-04	1.3E-04	NA	1.5E-04	1.6E-04
	StdDev	NA	4.6E-05	2.4E-05	NA	3.6E-05	5.9E-05
^{238}Pu Activity Concentration, Bq m^{-3}	N	0	3	0	0	1	2
	Mean	---	3.4E-09	---	---	1.5E-09	3.4E-09
	StdDev	---	3.3E-09	---	---	.	3.3E-09
^{238}Pu Activity Density, Bq g^{-1}	N	0	3	0	0	1	2
	Mean	---	9.4E-05	---	---	3.4E-05	8.3E-05
	StdDev	---	5.8E-05	---	---	.	4E-05
^{239,240}Pu Activity Concentration, Bq m^{-3}	N	32	50	10	27	48	43
	Mean	1.1E-08	1.8E-08	1.2E-08	8.1E-09	1.3E-08	1.4E-08
	StdDev	9.7E-09	1.3E-08	6.5E-09	5.5E-09	8.5E-09	7.9E-09
^{239,240}Pu Activity Density, Bq g^{-1}	N	32	50	11	27	49	45
	Mean	4.8E-04	5.5E-04	3.6E-04	4.8E-04	4.5E-04	3.5E-04
	StdDev	1.8E-04	2.3E-04	9.6E-05	2.9E-04	1.5E-04	1.0E-04

[a]N stands for number of samples with masses or activities above detection limits.

Table 7. Revised Aerosol Sampling Program for the WIPP EM (from March 2002 forward)

Sampler	Analytes	Frequency
High volume TSP	Mass & Radionuclides	Monthly
Graseby-Anderson Dichotomous	Trace Elements	Matched to TSP-HI VOL

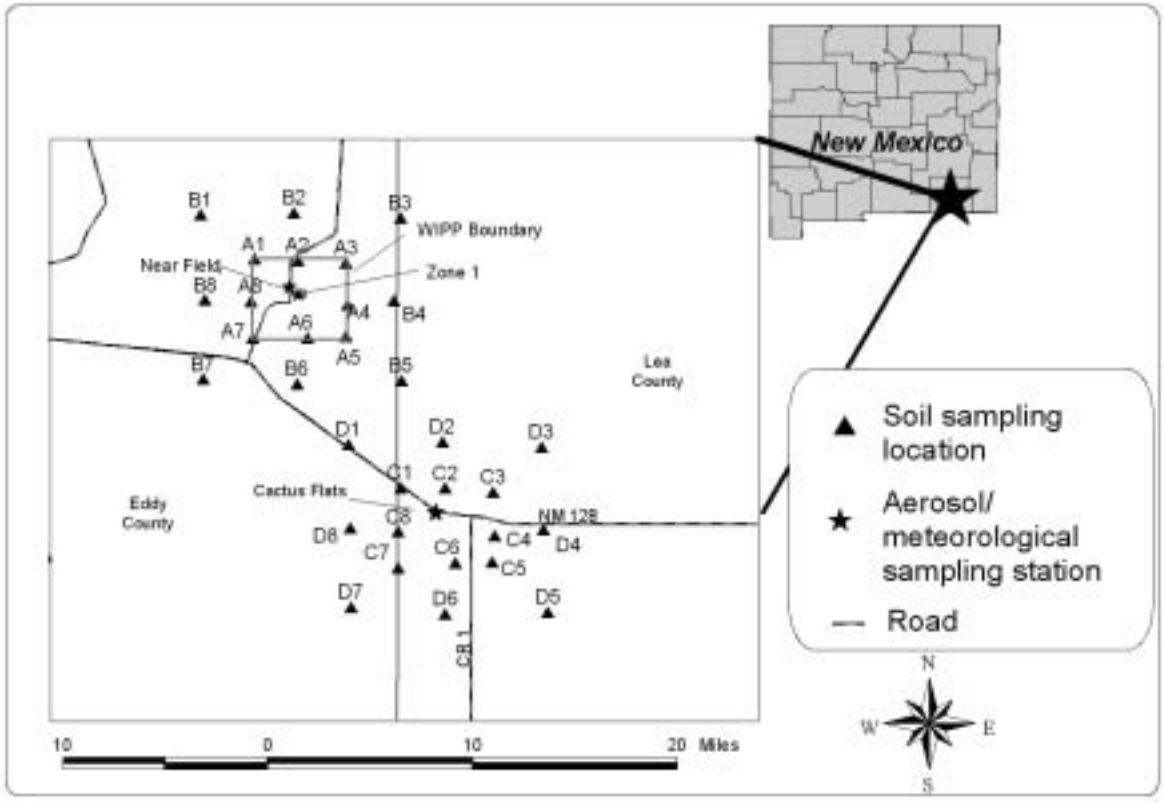

Figure 1. Sampling Locations in the Vicinity of the WIPP

Aerosol sampling monitoring is conducted at Near Field and Cactus Flats.
Meteorological monitoring is conducted at Cactus Flats.

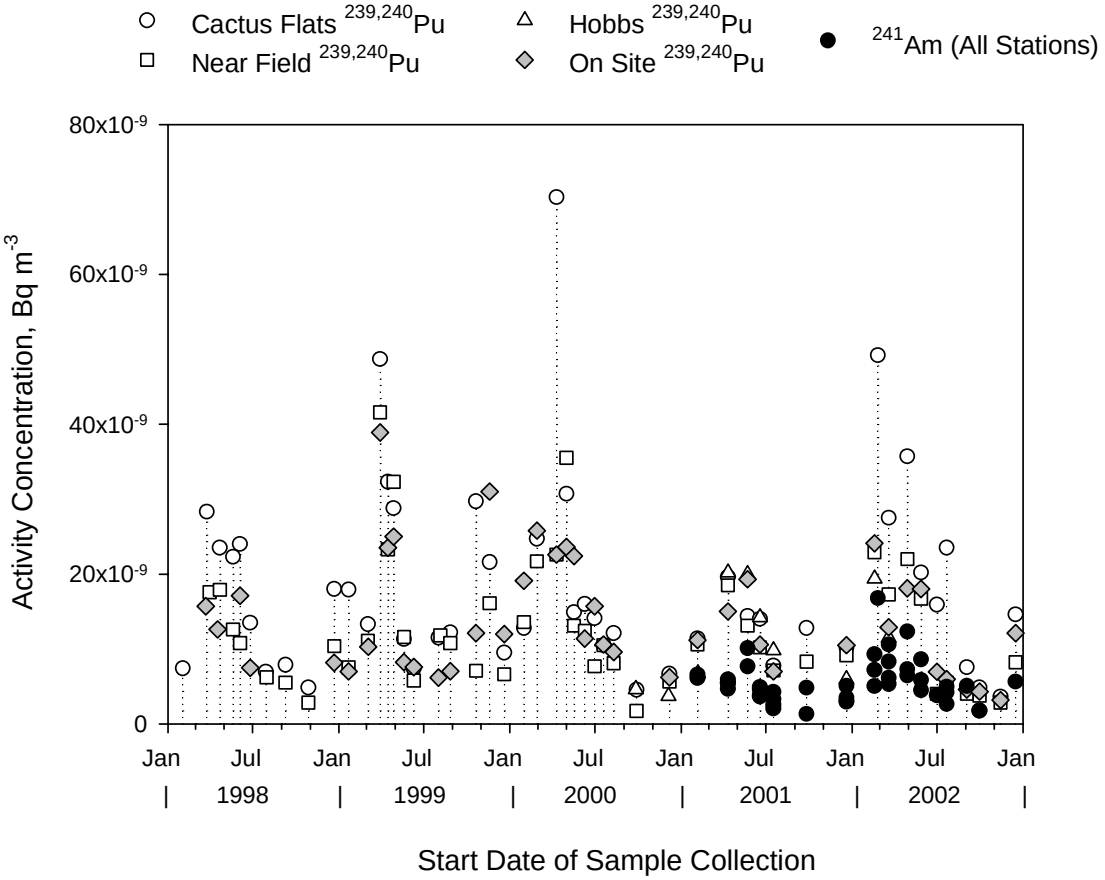

Figure 2. Timeseries plots for ^{239,240}Pu and ^{241}Am in high-volume aerosol samples from the Carlsbad/WIPP area.

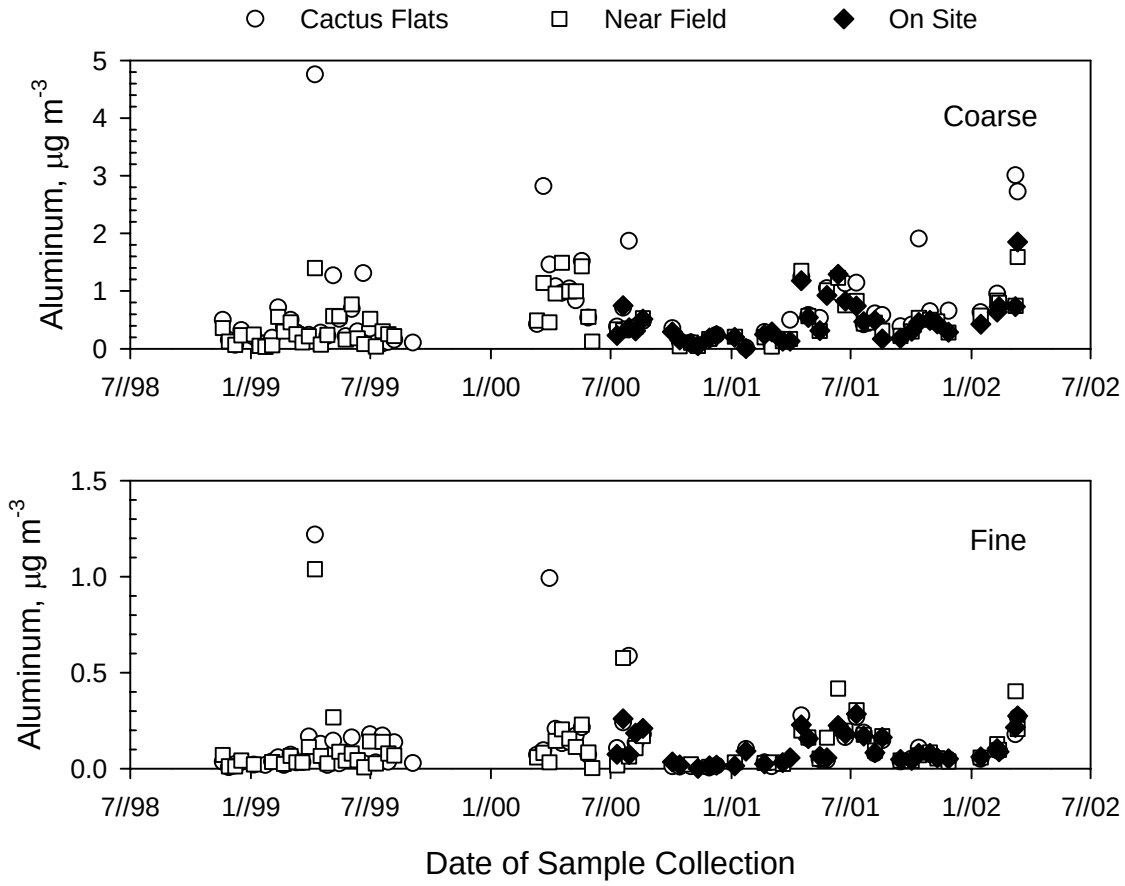

Figure 3. Timeseries plots for aluminum, an indicator of mineral dust, in the coarse and fine dichotomous aerosol sample fractions.

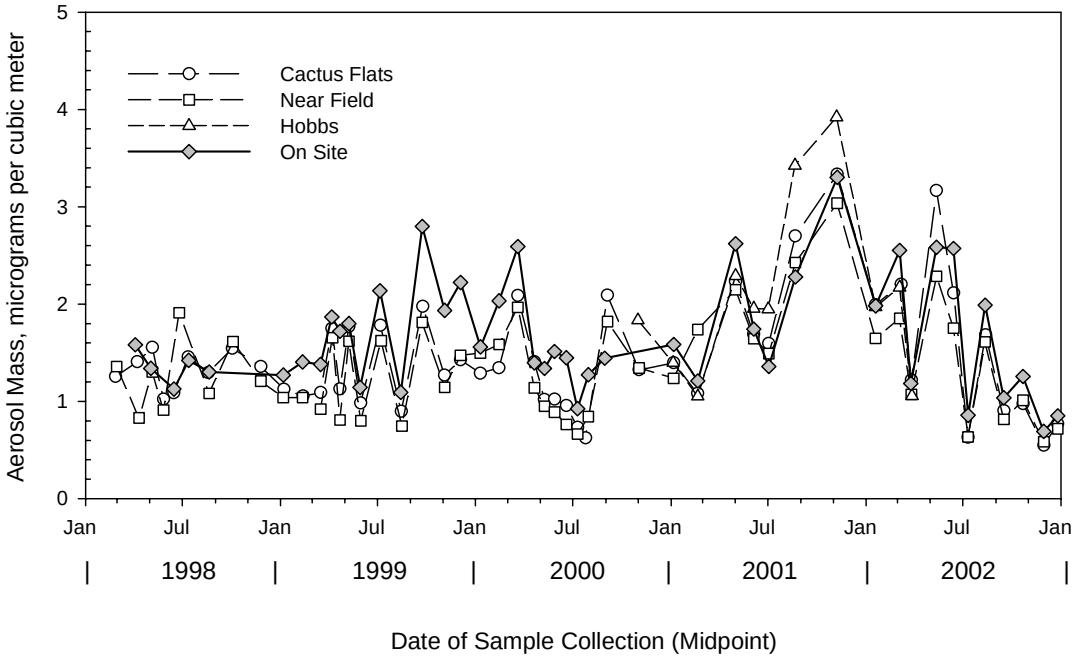

Figure 4. Timeseries plots for mass loadings in high-volume total suspended particle (TSP) aerosol samples.

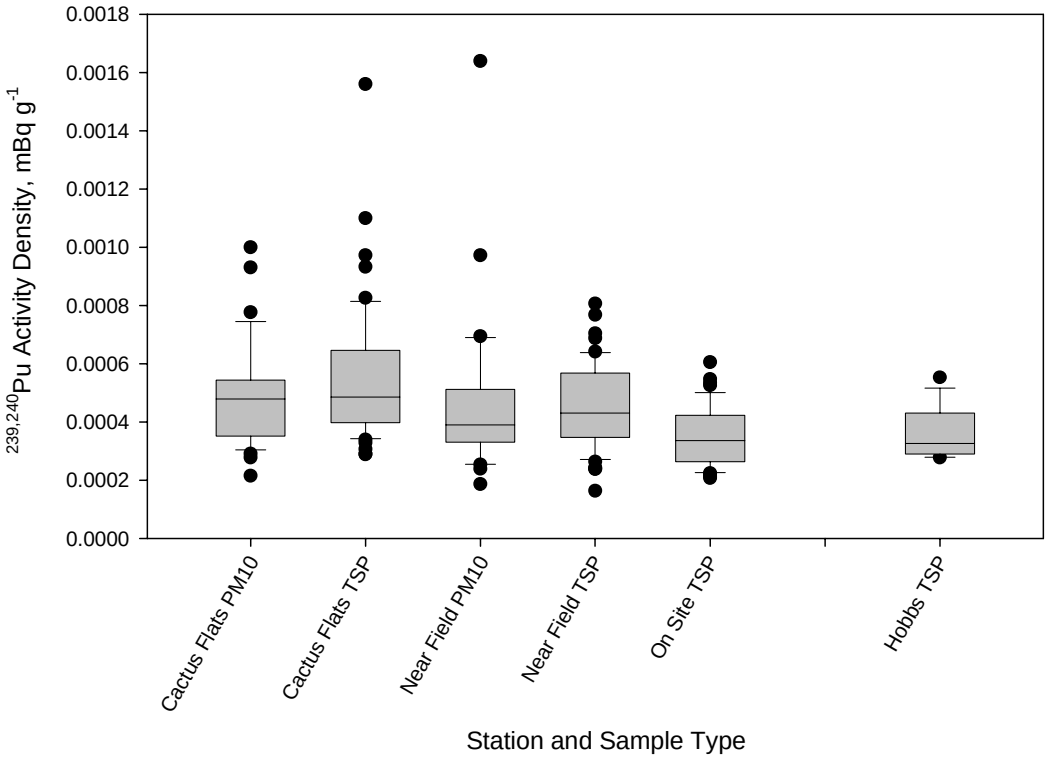

Figure 5. Box and whisker plots for ^{239,240}Pu activity densities in high-volume aerosol samples.

The edge of the box closest to zero indicates the 25th percentile, the line in the box marks the median, and the edge of the box farthest from zero indicates the 75th percentile. The whiskers above and below the box mark the 90th and 10th percentiles, respectively.

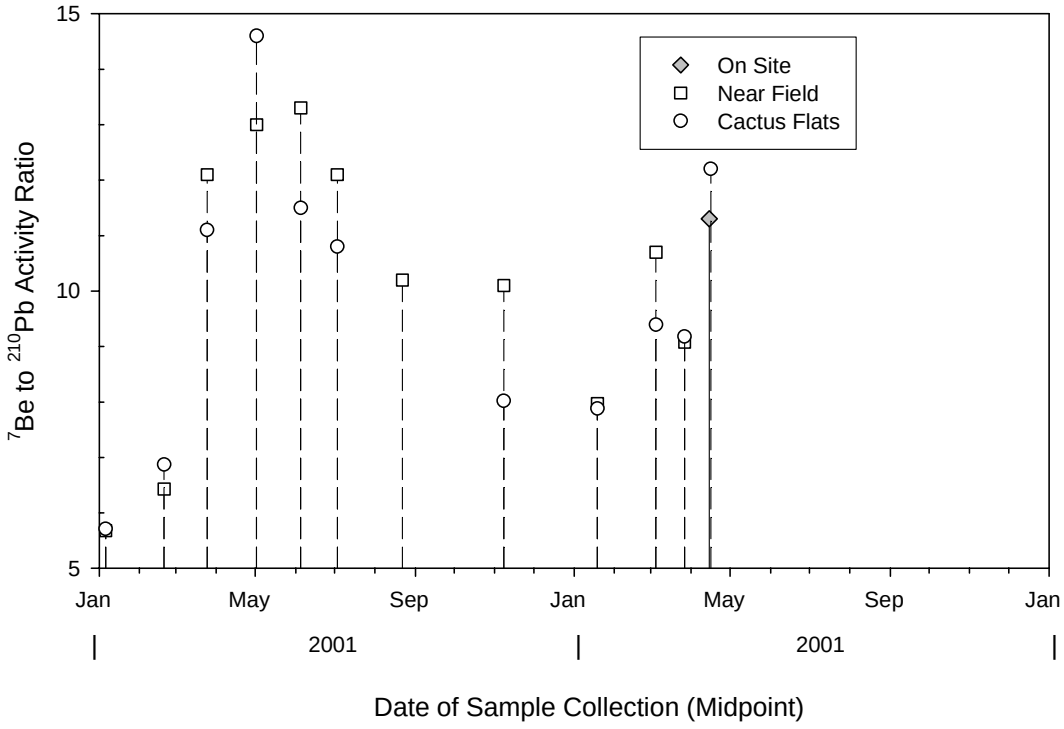

Figure 6. Timeseries plot of the ^{7}Be/^{210}Pb ratio in high-volume aerosol samples from the Carlsbad/WIPP area.

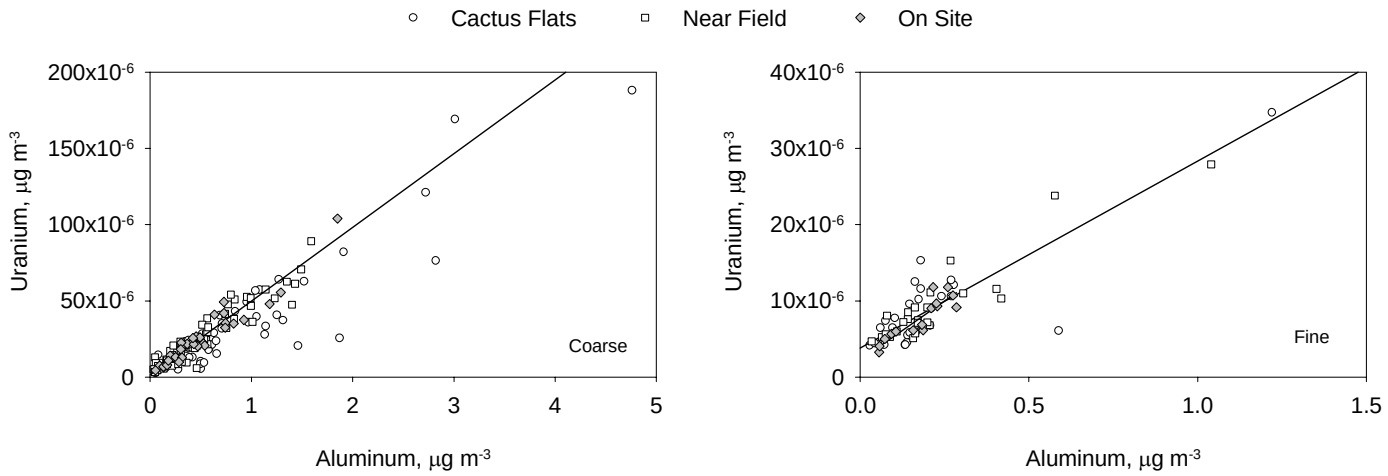

Figure 7a-b. Relationships between U vs. Al (an indicator of mineral dust) in coarse and fine aerosols.

Coefficients for U vs Al (7a, left): intercept = 1.41×10^{-6}, slope = 4.84×10^{-5}, $r^2 = 0.92$, (7b, right): intercept = 3.83×10^{-6}, slope = 2.45×10^{-5}, $r^2 = 0.85$

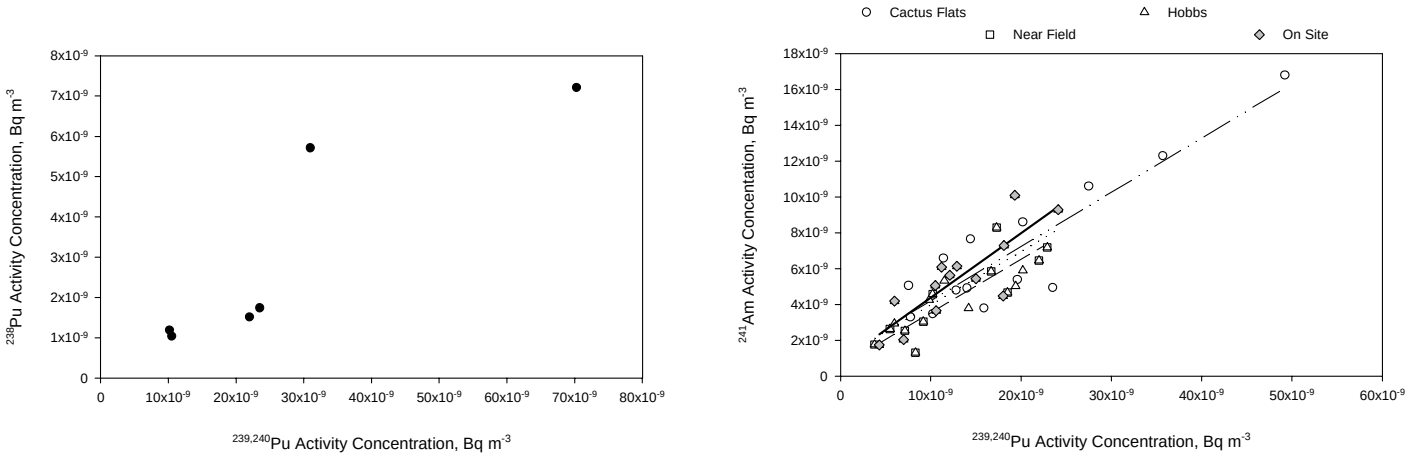

Figure 8a-b. Relationships between ^{238}Pu vs. ^{239,240}Pu (left) and ^{241}Am vs. ^{239,240}Pu (right).

Regression Lines for ^{241}Am vs. ^{239,240}Pu are similar for all stations (Cactus Flats = dash-dot-dot-dash; Near Field = dashed; Hobbs = dotted; On Site = solid, dark). The intercept for the ^{241}Am vs. ^{239,240}Pu regression for all stations combined was 9.7×10^{-10}; the slope was 0.31; and the coefficient of determination, r^2, was 0.78

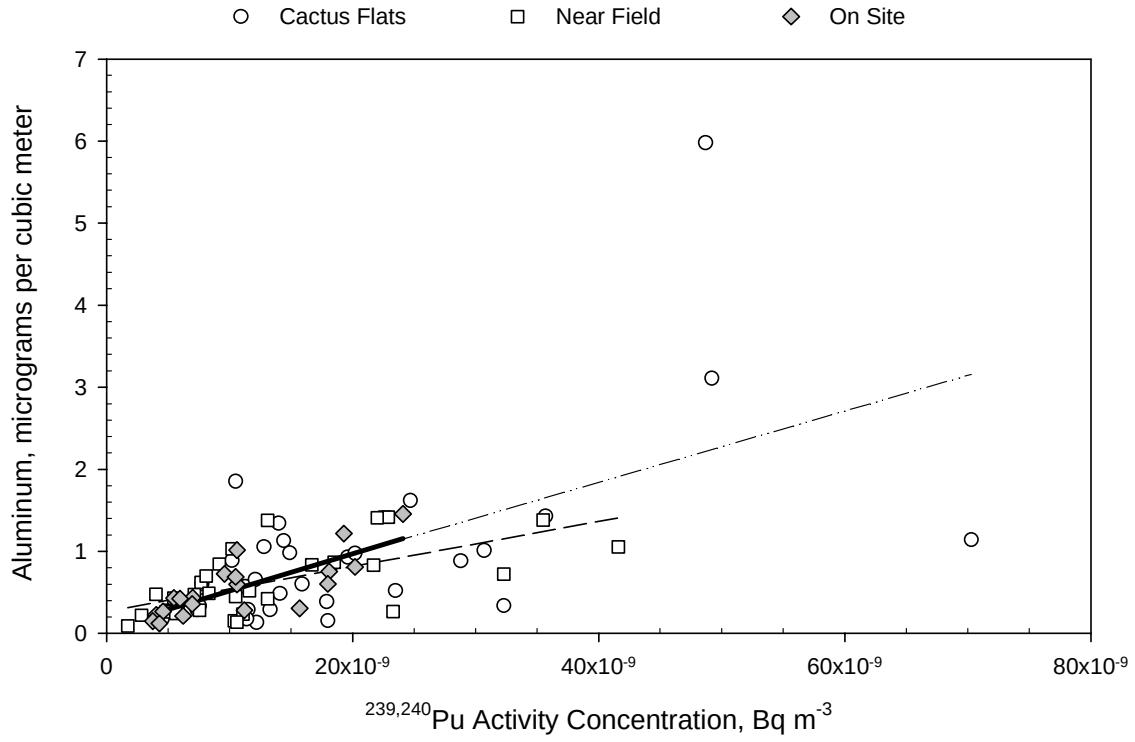

Figure 9. Relationship between Al (an indicator of mineral dust) vs. ^{239,240}Pu in aerosols.

The correlations between Al and ^{239,240}Pu for aerosols from all stations were significant at $p < 0.01$. The coefficients of determination $r^2 = 0.33$, 0.40 and 0.61 for Cactus Flats, Near Field, and On Site, respectively. (Regression lines: Cactus Flats = dash-dot-dot-dash; Near Field = dashed; On Site = solid, dark).

Development of Methods for Monitoring Volatile Organic Compounds

Introduction

A section of the permit governing the operation of the WIPP, i.e., New Mexico Administrative Code 4.1, Subpart V, § 264.602 and § 270.23, mandates the monitoring of nine volatile organic compounds (VOCs) in the WIPP underground to confirm that their respective concentrations of concern (COCs) are not exceeded. The COCs for these compounds for Drift E-300 of the WIPP underground range from 45 to 1930 ppbv (parts per billion by volume) according to Table N-2 of the final RCRA Permit. The nine contaminants of concern are listed in Table 8.

The current VOC monitoring program at the WIPP is based on the "*Confirmatory Volatile Organic Compound Monitoring Plan*" (Waste Isolation Pilot Plant, 2000) prepared by the Environmental Monitoring group of the WIPP management and operations contractor, WTS. That plan outlines procedures for the collection and analysis of VOC samples from two stations in the WIPP underground. The data from two stations, one upwind and the other downwind of Panel 1 in E-300 (which is where the first containers of waste have been emplaced), was designed to provide information on the quantities of VOCs released from the containers of mixed waste. Special quality assurance requirements for the VOC confirmatory monitoring were specified in "*Quality Assurance Project Plan for Confirmatory Volatile Organic Compound Monitoring*" (Waste Isolation Pilot Plant, 2001). At present, the VOC sampling operations are managed by WTS, and the analyses are performed by a contract laboratory.

Through a cooperative arrangement with LANL, the Environmental Chemistry Group of CEMRC has been given access to a Hewlett-Packard 6890/5973 (now Agilent Technologies) gas-chromatograph/mass spectrometer (GC/MS). This is a modern and sophisticated instrument well suited for VOC analyses. The GC/MS is being used to develop methods for VOC analyses, with the expectation that CEMRC will take over the VOC analyses for compliance monitoring. Progress made in terms of methods development for the VOC analyses is summarized herein.

Summary of VOC Analytical Methods Development

The methods described in the WTS "*Confirmatory Volatile Organic Compound Monitoring Plan*" mentioned above are based on the collection of ambient air samples in pressurized canisters. That document repeatedly refers to "concepts" described in TO-14A, and as a result the methods development at CEMRC focused on VOC sampling using specially prepared stainless steel canisters. There are in fact three relevant EPA approved methods for ambient atmospheric VOC sampling and analyses, these are:

1) EPA Air Analysis Compendium Method TO-14A "Sampling and Analysis of Ambient Air"

2) EPA Air Analysis Compendium Method TO-15 "Determination Of Volatile Organic Compounds (VOCs) In Air Collected In Specially-Prepared Canisters And Analyzed By Gas Chromatography/Mass Spectrometry (GC/MS)" and

3) EPA Air Analysis Compendium Method TO17 "Determination of Volatile Organic Compounds in Ambient Air Using Active Sampling Onto Sorbent Tubes."

In early 2002, information regarding vendors and pricing for equipment needed for VOCs analysis by EPA Method TO-14A was obtained. At that time, it was understood that the main items to be purchased were the canister cleaning system, some canisters, which were needed for the preparation of calibration standards, an ion gauge controller, and a cooled injection system (CIS) for the HP GC/MS system. The method also recommended a preconcentrator system for the samples, but that apparatus did not appear to be required by the method. Because it was not required and constituted what appeared to be an

unnecessary expense, the preconcentrator was not purchased.

In May, 2002, a Gerstel CIS4 CIS and an Agilent ion gauge controller were installed on the GC/MS system. Liquid samples of single analytes of interest, in addition to a liquid mixture containing all components analyzed by Method TO-14A were obtained, and tests run to evaluate the separation of components. It was determined that an analytical column more suited to VOCs analysis was needed, and so a Supelco SPB-624, 0.25mm X 60m column was procured and installed in July, 2002. Results from the analysis of liquid samples using that column demonstrated good system sensitivity and component separation.

Based on the success of those preliminary results, a series of tests of Silicosteel® tubing, a material being considered for use as a sample inlet, was performed using the GC/MS system in 2002. That was a collaborative study involving scientists from LANL and CEMRC. A final report of the Silicosteel® tests was scheduled for issue in 2003.

Briefly, the results of the Silicosteel® tests indicated that some of the VOCs may be retained in 500-foot lengths of the Silicosteel®tubing but the time delays for even the most strongly retained compounds are 30 minutes or less for flows designed to simulate sampling conditions in the underground. The delays in the appearance of the VOCs that were observed could largely be explained by volatilization artifacts caused by the sample introduction system of the test apparatus. Furthermore, the maximum degree of retention was short relative to the 12-hr intervals over which the WIPP underground samples would be collected.

Canister Cleaning System

A contract for the purchase of a canister cleaning system was awarded in early April 2002, and various parts of the cleaning system and canisters arrived in May and June of that year. The system was set up in early July, but it could not be made to function properly. The vendor was contacted numerous times regarding a variety of problems, but these problems were consistently attributed to operator error and unfamiliarity with the system. The system was returned to the vendor once for repair and when returned was still non-operational. In late August, a serious design flaw was discovered in the cleaning system (it would not operate at the reduced atmospheric pressure of Carlsbad), and this flaw was corrected by replacement of a pressure sensor. Thereafter, another design flaw was discovered and eventually the decision was made to return the original system and obtain one from an alternate source.

A second cleaning system was obtained from a different vendor, and it was installed in late November. This second system has functioned reliably, that is, without leaks or serious problems from the time of purchase until the present. The inability to clean canisters resulted in a long delay in preparation of calibration gas standards for method TO-14A. Following the set-up of the second canister cleaning system in the late fall/early winter of 2002, preparation of standards commenced, and attempts at calibrating the GC/MS system with actual gas standards began in earnest.

Calibration of the GC/MS

Early attempts demonstrated that the system sensitivity for compounds of interest was not within range of the required reporting limits (see Table 8 for reporting limits). Various methods for improving sensitivity were attempted. Great improvement was gained through the use of an inlet liner packed with glass beads and CarbotrapC®. The signals obtained from the use of this packed liner were sufficient to perform an actual calibration. Fig. 10 contains an example of a total ion chromatogram obtained using the current analysis method. A 600 mL injection of a 50 ppbv calibration standard was injected to obtain this chromatogram.

Methods development studies showed that some compounds in the calibration mixture could not be reliably separated and resolved by adjusting the chromatographic conditions (the CIS temperature and ramp, gas flow, oven temperature and ramp, etc). Through consultation with the current WTS vendor for the VOC analyses, it was determined that chromatographic separation of all components

was not necessary so long as there were no interferences of the ions used for quantitation.

Fig. 11 is an example of a total ion chromatogram showing an unresolved peak which contains both 1,1-Dichloroethene and 1,1,2 – Trichloro - 1,2,2 - trifluoroethane. Although some peaks for these compounds co-elute, they can still be quantitated based on different primary ions. Since their mass spectra do not contain common target ions, there are no interferences due to the lack of chromatographic resolution. Fig. 12 shows two overlaid single-ion chromatograms. The single-ion chromatogram of mass 61 represents the target ion for 1,1-Dichloroethene quantitation while the single ion chromatogram of mass 151 represents the target ion for 1,1,2-Trichloro-1,2,2-trifluoroethane. The separation of the unresolved peak into two single-ion peaks demonstrates that in GC/MS analysis, a combination of retention times and ion mass/charge separation can sometimes be used to identify and quantify VOCs of concern. The methods for identifying and quantifying the VOCs will be updated based these results.

Next Steps

Despite the progress made toward calibrating the GC/MS, continuing calibrations of the instrument were not consistently successful; that is, the results were not within specifications for all compounds of interest. This lack of consistency showed that the system as currently configured did not have the requisite sensitivity and repeatability for reliable routine VOC analyses. From discussions with other operators performing similar analyses and through investigations of existing GC/MS systems, it became evident that a different sample introduction system would likely be required to attain the required performance. Various options for sample preconcentration were being evaluated at the time this report was prepared.

In addition to equipment upgrades, training is scheduled for the main operator of the Agilent GC/MS system; this will mainly involve participation in a course given by the instrument's manufacturer on "Environmental GC-MSD Instrument and ChemStation Operation". Finally, experienced analysts who work with VOCs as well as vendors who make the equipment used for these analyses will continue to consulted to optimize the instrumentation and to improve the analytical methods.

Table 8. Volatile Organic Compounds of Concern

	Compound	Formula	Laboratory Reporting Limit, ppbv	CAS No.
(1)	Carbon tetrachloride	CCl_4	2	56-23-5
(2)	Chlorobenzene (monochlorobenzene, benzene chloride)	C_6H_5Cl	2	108-90-7
(3)	Chloroform (trichloromethane)	$CHCl_3$	2	67-66-3
(4)	1,1-Dichloroethylene (vinylidene chloride, 1,1-DCE)	$C_2H_2Cl_2$	2	75-35-4
(5)	1,2-Dichloroethane (1,2-DCA, DCA, ethylene dichloride, EDC, sym-dichloroethane)	$C_2H_4Cl_2$	2	107-06-2
(6)	Methylene chloride (dichloromethane)	CH_2Cl_2	5	75-09-2
(7)	1,1,2,2-Tetrachloroethane (sym-tetrachloroethane)	$C_2H_2Cl_4$	2	79-34-5
(8)	Toluene (methylbenzene)	C_7H_8	5	108-88-3
(9)	1,1,1-Trichloroethane (methyl chloroform)	$C_2H_3Cl_3$	5	71-55-6

Table 9. Initial Calibration Summary

Initial Calibration Relative Response Factors[a]							
Compound	**50ppbv**	**20ppbv**	**10ppbv**	**5ppbv**	**2ppbv**	**Average**	**%RSD**
Chloroform	3.326	3.06	2.51	3.624	2.601	3.024	15.65
1,1-Dichloroethene	1.619	1.589	1.451	1.85	1.318	1.566	12.73
1,2-Dichloroethane	2.038	1.893	1.787	2.033	1.412	1.832	14.05
Methylene Chloride	0.782	0.984	1.108	1.107	1.012	0.999	13.34
Carbon Tetrachloride	0.83	0.839	0.636	1.014	0.672	0.798	18.94
1,1,1-Trichloroethane	0.712	0.703	0.594	0.846	0.552	0.682	16.9
Chlorobenzene	2.157	2.104	1.879	2.289	1.642	2.014	12.67
1,1,2,2-Tetrachloroethane	1.403	1.425	0.864	0.872	0.554	1.024	37.0
Toluene	2.572	2.033	1.625	2.093	1.346	1.934	24.3

[a]Relative response factors at each level are the average of response factors from two runs. Relative response factors are the number of raw counts for the compounds of interest relative to the counts for a simultaneously run internal standard

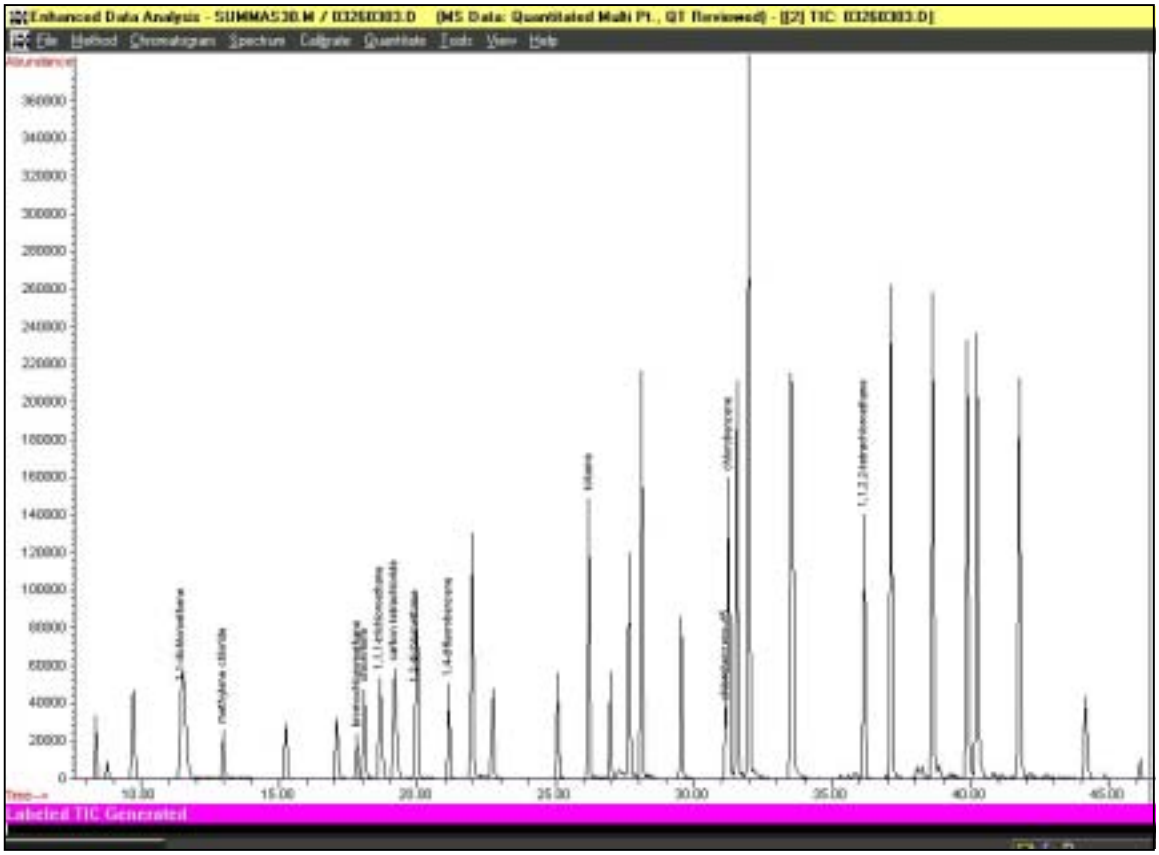

Figure 10. Total ion chromatogram of a 50 ppbv calibration standard mixture.

Note: The calibration standard used to produce this chromatogram contains all 39 components normally analyzed for EPA Method TO-14A. Only those components of interest listed in Table 8 are labeled.

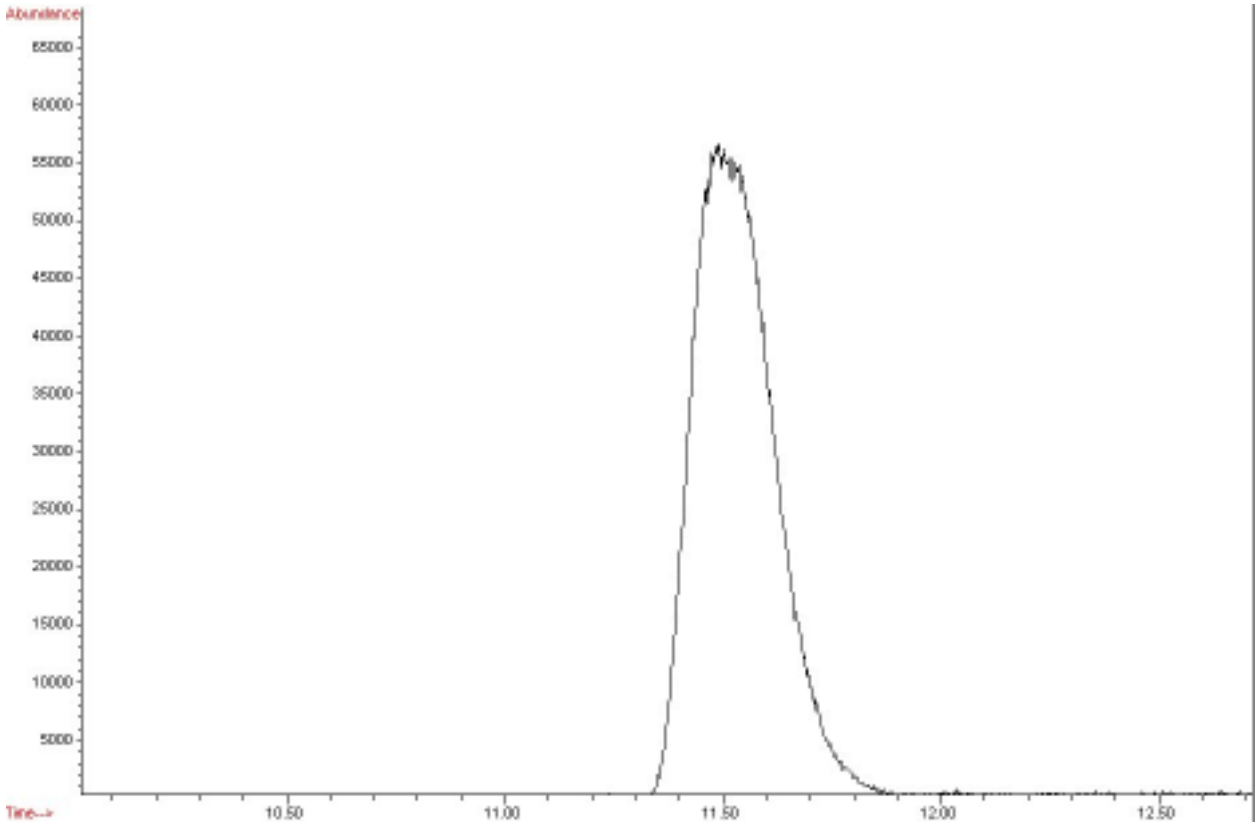

Figure 11. Total ion chromatogram showing unresolved peak

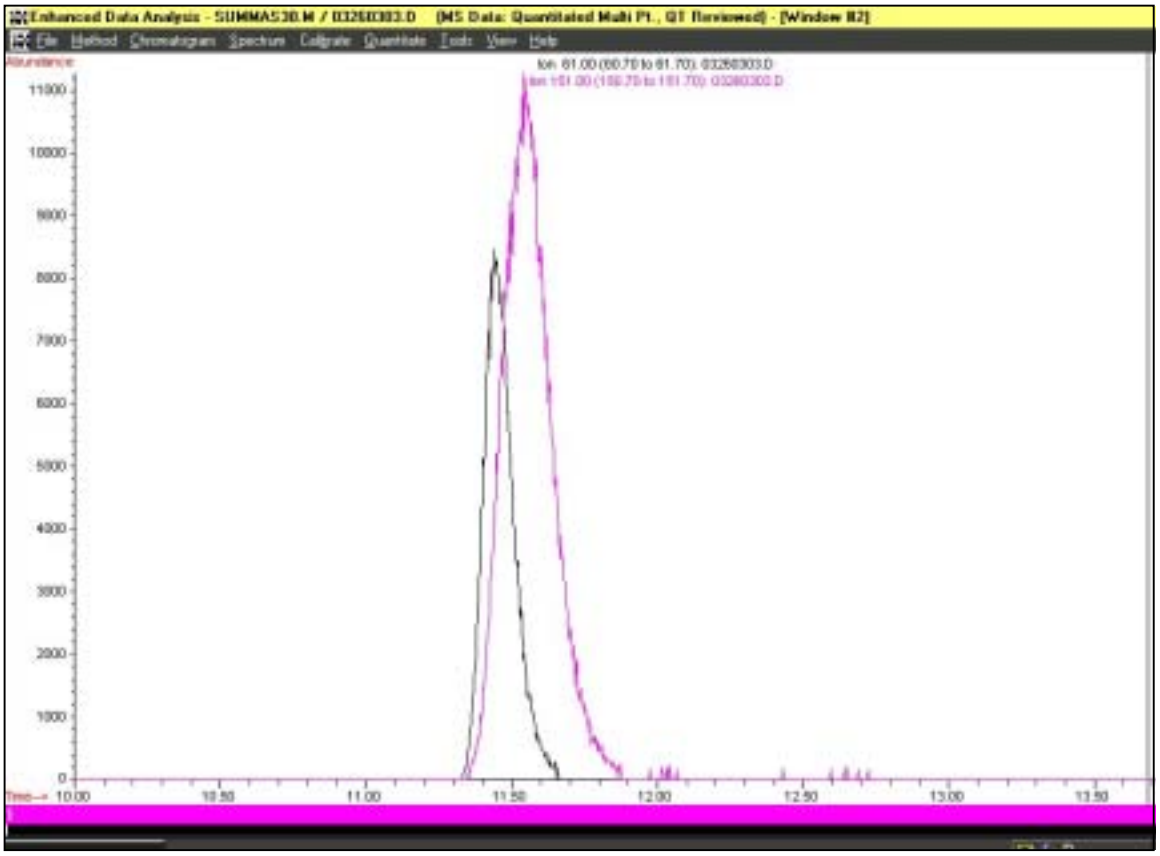

Figure 12. Single ion chromatograms of the peak from Figure 11.

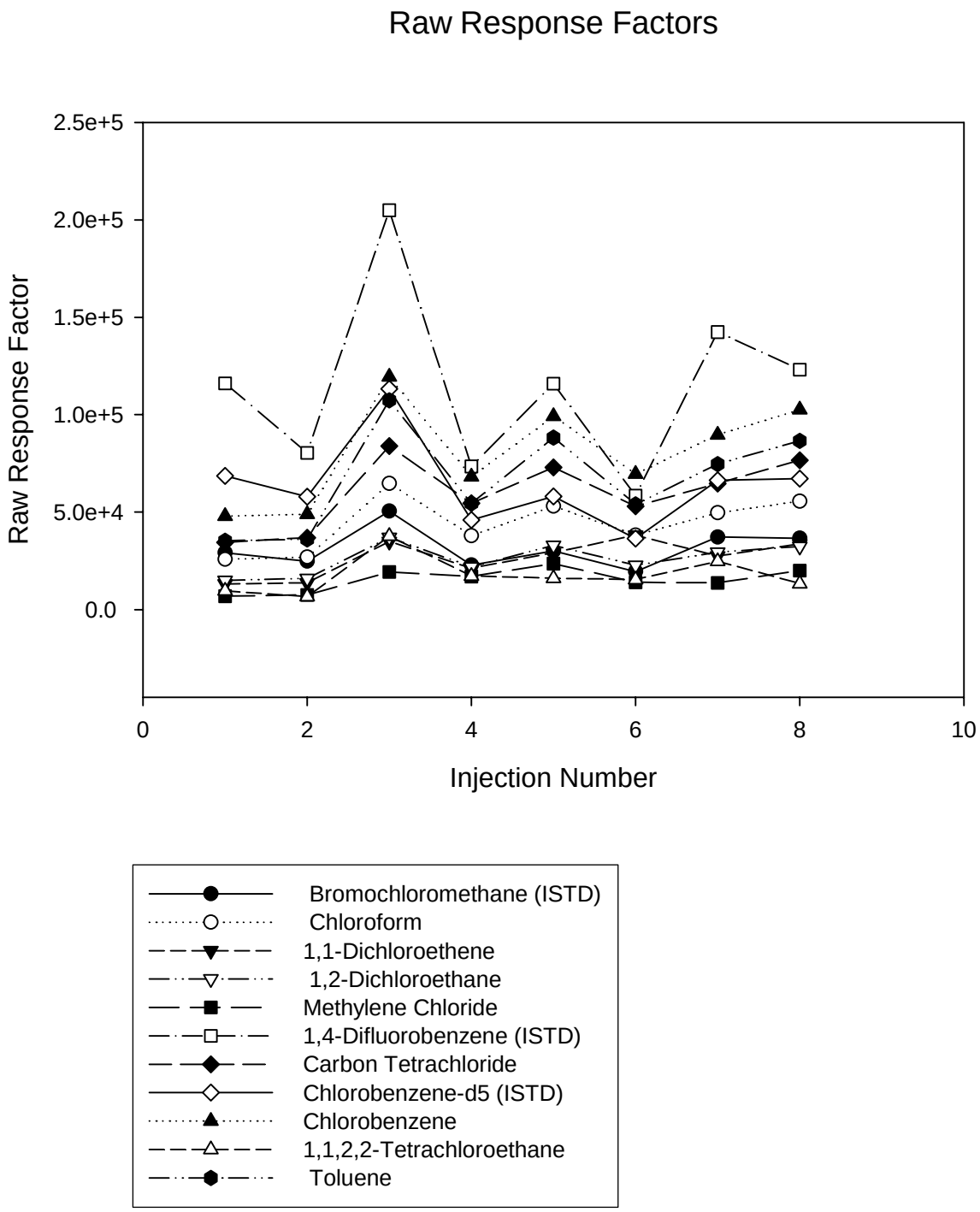

Figure 13. Variations in response factors for VOCs of interest and internal standards (ISTD).

Radionuclides and Inorganics in WIPP Exhaust Air

Introduction

Beginning in late 1998, studies have been conducted on the loadings and composition of aerosol samples collected from the WIPP exhaust shaft. The samples for this part of the WIPP EM program are collected at Station A, which is an aerosol sampling platform shared with the Environmental Evaluation Group (EEG) and Washington TRU solutions (WTS). Data from Station A not only provide a means for monitoring the effluent from the WIPP, but data from the station also could be of critical importance for reconstructing public or worker exposure if a release were to occur at the WIPP. This report summarizes analyses completed through the 3rd Quarter (Sept. 30th) 2002.

From a practical standpoint, the WIPP exhaust shaft is where radioactive or hazardous materials would most likely first be detected in the event of a release. Therefore, CEMRC has developed procedures and methods to provide information about the exhaust emissions on a short turn-around time schedule (i.e., weeks). This addresses a strategic need for the monitoring program because most of the other WIPP EM analyses require relatively long turn-around times; that is, it may take several months to complete the chemical and radiochemical analyses after the samples are collected. Therefore, the Station A studies provide quick-look monitoring results, which while less specific and less detailed than those from other studies, can used to trigger more detailed investigations if necessary. This was dramatically demonstrated in January 2001 when a fire extinguisher discharged, and the CEMRC monitoring program identified elevated gross beta radioactivity from the extinguisher on Station A filters.

Methods

The aerosol samples at Station A are typically collected daily except for weekends (the weekend samples run from Friday to Monday so the coverage is continuous). If waste operations occur on the weekend then samples are also collected on the weekends. The samples are collected from a shrouded probe, commonly referred to as a fixed air sampler (FAS), which has a transfer line running to each of three sampling legs; thus a total of three concurrent samples can be collected from the FAS, one each for CEMRC, EEG and WTS.

Detailed descriptions of the sampling and analytical methods have been included in the CEMRC Annual Reports for 1999 and 2000. Briefly, after the samples are returned to the laboratory, the individual filters are weighed to determine mass loadings, and they are counted for gross alpha/beta activities. Elemental and gamma-ray analysis are conducted on weekly composites of the filter, and quarterly composites are used for the determination of actinide activities.

As with all other WIPP EM activities, the Station A studies have come under more rigorous quality control in FY 2002.

Results and Discussion

Routine Monitoring

The Station A studies are unique among the WIPP EM activities in that quick-look data sets are posted for public inspection at the CEMRC website (http://www.cemrc.org) within a few weeks after the samples are collected. The strategic design for the WIPP EM was to compare pre- vs. post-disposal data. The first radioactive waste shipments were received at the WIPP on March 26, 1999, and this is considered the cut-off between the pre-disposal vs. operational phase.

Operational values of gross alpha activity concentration and density (Table 10) ranged from < MDC (≈0.1 mBq m^{-3}) to 0.42 mBq m^{-3} and < MDC (≈0.7 Bq g^{-1}) to 9.6 Bq g^{-1}, respectively. Values of gross beta activity concentration and density ranged from < MDC (≈ 0.2 mBq m^{-3}) to 58 mBq m^{-3} and < MDC (≈ 1.7 Bq g^{-1}) to 190 Bq g^{-1}, respectively. In general, values of gross alpha and beta concentration and density are either unchanged or have decreased since the WIPP

began receiving waste in March 1999. (Table 10, Figs. 14 and 15).

The WIPP began receiving mixed waste on September 9, 2000, and therefore for the elemental constituents, samples collected prior to that date represent a baseline while the results for samples collected afterwards represent operational monitoring data. Numerous elemental constituents were quantified in weekly composites (Table 11). More than 85% of all of the 36 elements that can be routinely quantified with the ICP-MS used for the FAS sample analyses were observed in 75-100% of the weekly composites.

Many of the potentially toxic elements (e.g. Pb, Be, Cd, etc.) which are known or expected to be contained in WIPP mixed waste were already present in measurable amounts in the WIPP aerosol effluent prior to the receipt of mixed waste. The Station A studies show that the concentrations of the hazardous metals and other elements were highly variable even in the samples collected prior to receipt of the mixed waste.

Aside from the heavy metals, one non-radiogenic constituent of considerable interest is Na; this is because a variety of operations in the underground cause the generation of salt dusts. The Na concentration in the FAS samples showed strong temporal peaks, and these generally appeared when mining or other activities (e.g. panel excavation) were occurring in the repository (Fig.16).

Another non-radiogenic constituent of interest is Al. The Al concentrations in FAS samples exhibit a seasonal cycle, peaking in the spring (Fig. 17). As noted in the chapter of this report dealing with ambient aerosols, this phenomenon has also been observed in ambient aerosol samples collected above ground in the vicinity of the WIPP. Spring is typically the windiest time of the year and the increase in Al concentrations in the exhaust air likely is caused by an increase in dust (soil) entrained in the ambient air entering the mine air intake and circulated throughout the mine. Aluminum has been used as an indicator of mineral dust, and a relationship has been demonstrated between Al, and ^{239,240}Pu concentration in ambient aerosols (Arimoto et al., 2002) and soil (Kirchner et al., 2002) where an ambient aerosol ^{239,240}Pu:Al ratio of approximately 0.025 nBq ng^{-1} has been observed.

Assuming the ^{239,240}Pu:Al ratio of 0.025 nBq ng^{-1} observed on the surface holds true for aerosols entering the WIPP underground, One can use this information to address the question – will we ever detect plutonium in the exhaust air samples? The concentration of ^{239,240}Pu in the WIPP underground air can be estimated using the Al concentration observed in the exhaust air by:

$$[Pu] = R*[Al] \qquad \text{Equation (1)}$$

Where:
[Pu] = ^{239,240}Pu concentration (nBq m^{-3})
R = ^{239,240}Pu to Al ratio = 0.025 nBq ng^{-1}
[Al] = Exhaust air Al concentration (ng m^{-3})

If the average (450 ng m^{-3}) and maximum (1400 ng m^{-3}) Al concentrations measured during the operational monitoring period (Table 11) are inserted in equation 1, the ^{239,240}Pu concentration in WIPP underground air is estimated to range from approximately 11.3 to 35 nBq m^{-3}. This estimate is very close to the detection limit of the composite samples analyzed for ^{239,240}Pu by alpha spectroscopy (Table 12). To date, all quarterly composite samples analyzed by alpha spectroscopy have been below the MDC for ^{239,240}Pu with the MDC's for the individual quarterly samples ranging from 27 to 137 nBq m^{-3} (average = 75 nBq m^{-3}).

Since the MDCs for ^{239,240}Pu are only approximately a factor of 2-3 higher than the predicted concentrations, it appears very probable that ^{239,240}Pu could be detected in future composite samples with source being dust-loaded ambient air circulating through the underground as opposed to a release of Pu from WIPP operations. This is important to note because while it may not be possible to definitively prove that ambient aerosols are the source of Pu detected in an exhaust sample, it at least provides a defensible explanation of the source of Pu.

This is further supported by the ^{7}Be data. With the exception of ^{7}Be, no detectable gamma-emitting radionuclides were observed during this monitoring period (June 2001 –

September 2002). ^{7}Be is a short-lived radionuclide ($T_{1/2}$ = 53 days) that is produced in the stratosphere through spallation of atmospheric gases (not occurring naturally in the WIPP underground). ^{7}Be was detected in approximately 52% of samples, ranging in activity concentration and density from 3 to 13 mBq m^{-3} and 12 to 332 Bq g^{-1}, respectively. For detectable results, mean values (± SE) of activity concentration and density were 6.7 (± 0.4) mBq m^{-3} and 85 (± 15) Bq g^{-1}, respectively. ^{7}Be values during this monitoring period were consistent with those reported in the CEMRC 1999, 2000 and 2001 Reports. The presence of ^{7}Be in the exhaust is also an indicator that the aerosols entering through the WIPP air intake eventually reach the exhaust system and are released as exhaust effluents. This finding supports the hypothesis that aerosols containing radionuclides (e.g. ^{239,240}Pu, ^{137}Cs) enter the underground via the ventilation system, circulate and are expelled in the exhaust where they may be detected in FAS samples even in the absence of a WIPP-related contamination event.

Isotopes of naturally occurring U and Th isotopes have been detected in all quarterly composites reported for this monitoring period. Concentrations and activity densities of U and Th isotopes for all quarters reported herein were consistent with that reported in the CEMRC 1999, 2000 and 2001 Reports.

As stated above, no detectable concentrations of ^{238}Pu, ^{239,240}Pu, ^{241}Pu, or ^{241}Am (Table 12) were observed in any operational quarter reported herein. These results are consistent with those reported in the CEMRC 1999, 2000 and 2001 Reports.

Table 10. Summary Statistics for Gross Alpha/Beta Analyses of Daily FAS Filters

Gross Emission	[a]N	% ≥ [b]MDC	Activity Concentration ($Bq\ m^{-3}$)			Activity Density ($Bq\ g^{-1}$)		
			[c]Mean	[d]SE	[e]Max	Mean	SE	Max
Pre-Operations Baseline								
Alpha	71	100	3.1E-04	3.1E-05	1.5E-03	3.6E+00	5.8E-01	3.7E+01
Beta	71	100	1.1E-03	9.1E-05	4.9E-03	1.4E+01	1.9E+00	1.2E+02
Operational Monitoring April 1999 – June 2002								
Alpha	1378	50	9.4E-05	2.1E-06	4.2E-04	1.2E+00	4.6E-02	9.6E+00
Beta	1378	96	1.0E-03	5.2E-05	5.8E-02	1.1E+01	4.4E-01	1.9E+02

[a]N = number of samples
[b]MDC = minimum detectable concentration
[c]Mean = arithmetic mean
[d]SE = standard error
[e]Max = maximum

Table 11. Summary Statistics for Elemental Constituents in Weekly FAS Composites

Analyte	[a]Baseline Aerosol Concentration December 1998 – September 2000 (ng m^{-3})				[b]Operational Aerosol Concentration September 2000 – September 2002 (ng m^{-3})			
	[c]FD (%)	[d]Mean	[e]SE	[f]Max	FD (%)	Mean	SE	Max
Ag	79	1.1E-01	2.2E-02	1.2E+00	51	4.0E-01	1.4E-01	5.5E+00
Al	99	7.2E+02	9.7E+01	7.8E+03	100	4.5E+02	2.5E+01	1.4E+03
As	70	1.3E+00	1.2E-01	4.7E+00	76	1.1E+00	1.3E-01	9.1E+00
Ba	100	7.6E+00	4.1E-01	2.0E+01	100	6.3E+00	5.8E-01	5.2E+01
Be	7	2.9E-01	1.4E-01	9.8E-01	3	4.2E-01	1.1E-01	6.2E-01
Ca	100	4.1E+03	8.8E+02	7.0E+04	100	5.9E+03	1.4E+03	1.3E+05
Cd	82	9.4E-01	2.4E-01	1.5E+01	36	6.5E-01	1.6E-01	5.6E+00
Ce	99	7.4E-01	4.7E-02	2.1E+00	100	5.8E-01	4.2E-02	2.2E+00
Co	94	3.1E+00	4.5E-01	2.4E+01	98	7.2E+00	1.1E+00	8.5E+01
Cr	61	5.8E+01	1.3E+01	6.2E+02	93	8.5E+01	6.6E+00	4.3E+02
Cu	100	3.7E+01	2.0E+00	1.1E+02	79	4.9E+01	6.3E+00	3.3E+02
Dy	100	4.6E-02	3.4E-03	1.6E-01	44	6.8E-02	1.1E-02	4.9E-01
Er	96	2.8E-02	2.4E-03	1.5E-01	24	6.2E-02	1.3E-02	3.4E-01
Eu	93	1.5E-02	9.3E-04	3.7E-02	14	7.5E-02	3.3E-02	4.8E-01
Fe	100	8.0E+02	7.9E+01	5.8E+03	100	8.0E+02	5.6E+01	4.6E+03
Gd	99	7.2E-02	5.5E-03	2.8E-01	78	9.4E-02	9.4E-03	5.4E-01
Hg	30	1.6E-01	2.9E-02	5.7E-01	13	1.3E+00	2.6E-01	3.6E+00
K	99	1.3E+03	1.1E+02	5.4E+03	96	1.2E+03	1.1E+02	8.9E+03
La	100	4.5E-01	3.0E-02	1.3E+00	97	3.0E-01	1.8E-02	1.1E+00
Li	76	2.4E+00	2.7E-01	1.4E+01	70	1.7E+00	4.0E-01	2.7E+01
Mg	100	2.7E+03	6.4E+02	5.1E+04	100	2.1E+03	6.5E+02	6.1E+04
Mn	100	3.4E+01	3.5E+00	1.4E+02	100	3.1E+01	2.7E+00	2.2E+02
Mo	70	4.3E+00	1.3E+00	7.5E+01	96	8.6E+00	5.5E-01	2.6E+01
Na	99	6.7E+04	7.2E+03	2.9E+05	100	3.7E+04	3.5E+03	2.2E+05
Nd	100	3.1E-01	2.0E-02	9.2E-01	75	2.7E-01	1.9E-02	9.4E-01
Ni	91	1.8E+01	5.7E+00	4.2E+02	100	4.6E+01	4.1E+00	2.7E+02
Pb	100	7.0E+00	7.4E-01	4.6E+01	100	6.3E+00	8.0E-01	7.9E+01
Pr	100	8.9E-02	5.6E-03	2.7E-01	97	7.2E-02	6.5E-03	5.1E-01
Sb	100	3.1E+01	3.1E+00	2.2E+02	100	1.1E+01	7.6E-01	4.9E+01
Se	28	5.5E-01	4.7E-02	1.1E+00	19	6.0E-01	6.5E-02	1.3E+00
Sm	100	4.7E-01	2.0E-02	1.2E+00	47	4.8E-01	2.0E-02	7.9E-01
Sr	100	6.8E+01	1.8E+01	1.4E+03	100	1.0E+02	3.2E+01	3.1E+03
Th	94	1.1E-01	9.7E-03	4.8E-01	88	8.0E-02	9.1E-03	6.9E-01
Ti	99	4.4E+01	3.3E+00	2.1E+02	71	2.5E+01	3.0E+00	1.8E+02
U	91	4.7E-02	4.5E-03	2.4E-01	88	4.1E-02	5.2E-03	4.1E-01
Zn	98	2.7E+02	8.0E+01	4.7E+03	100	1.2E+02	1.5E+01	1.2E+03

[a]A total of 102 weekly composites were analyzed during this interval
[b]A total of 90 weekly composites were analyzed during this interval
[c]FD = frequency of detection
[d]Mean = arithmetic mean
[e]SE = standard error
[f]Max = maximum

Table 12. Results of Actinide Analyses for Quarterly FAS Composite Samples

Radionuclide	Activity Concentration ($Bq\ m^{-3}$)			Activity Density ($Bq\ g^{-1}$)		
	[a]C	[b]SD	[c]MDC	C	SD	MDC
Pre-Operational Baseline						
^{241}Am	<MDC	[d]NA	4.0E-08	<MDC	NA	4.3E-04
^{238}Pu	<MDC	NA	3.3E-08	<MDC	NA	3.5E-04
$^{239,240}Pu$	<MDC	NA	2.7E-08	<MDC	NA	2.9E-04
(e) ^{241}Pu	[f]NR	NR	NR	NR	NR	NR
^{228}Th	8.1E-07	5.6E-08	5.8E-08	8.7E-03	6.0E-04	6.2E-04
^{230}Th	7.5E-07	5.3E-08	6.5E-08	8.1E-03	5.7E-04	7.0E-04
^{232}Th	5.2E-07	4.0E-08	3.8E-08	5.6E-03	4.3E-04	4.0E-04
^{234}U	9.5E-07	5.3E-08	1.3E-08	1.0E-02	5.7E-04	1.3E-04
^{235}U	4.4E-08	1.6E-08	3.7E-08	4.8E-04	1.7E-04	3.9E-04
^{238}U	9.1E-07	5.2E-08	1.6E-08	9.8E-03	5.6E-04	1.8E-04
Operational Monitoring Second Quarter 1999 – Second Quarter 2001						
^{241}Am	<MDC	NA	6.9E-08	<MDC	NA	4.0E-04
^{238}Pu	<MDC	NA	1.1E-07	<MDC	NA	7.3E-04
$^{239,240}Pu$	<MDC	NA	8.9E-08	<MDC	NA	4.9E-04
^{241}Pu	<MDC	NA	2.2E-05	<MDC	NA	7.9E-02
^{228}Th	8.2E-07	3.9E-07	1.7E-07	5.8E-03	4.6E-03	1.0E-03
^{230}Th	5.7E-07	3.3E-07	1.4E-07	3.5E-03	2.0E-03	8.3E-04
^{232}Th	3.9E-07	2.0E-07	8.0E-08	2.6E-03	2.1E-03	4.9E-04
^{234}U	8.2E-07	3.5E-07	8.3E-08	5.2E-03	2.7E-03	5.7E-04
^{235}U	1.1E-07	6.7E-08	9.4E-08	7.0E-04	5.8E-04	5.7E-04
^{238}U	6.6E-07	3.2E-07	9.0E-08	4.1E-03	2.1E-03	6.1E-04
Operational Monitoring Third Quarter 2001						
^{241}Am	<MDC	NA	8.8E-08	<MDC	NA	1.3E-03
^{238}Pu	<MDC	NA	1.2E-07	<MDC	NA	1.8E-03
$^{239,240}Pu$	<MDC	NA	1.1E-07	<MDC	NA	1.6E-03
^{241}Pu	<MDC	NA	3.2E-05	<MDC	NA	4.7E-01
^{228}Th	5.2E-07	9.4E-08	2.0E-07	7.6E-03	1.4E-03	3.0E-03
^{230}Th	4.3E-07	7.9E-08	1.6E-07	6.2E-03	1.2E-03	2.4E-03
^{232}Th	3.4E-07	6.0E-08	1.1E-07	4.9E-03	8.6E-04	1.6E-03
^{234}U	8.5E-07	8.2E-08	8.1E-08	1.2E-02	1.2E-03	1.2E-03
^{235}U	<MDC	NA	1.1E-07	<MDC	NA	1.5E-03
^{238}U	5.4E-07	6.8E-08	1.1E-07	7.8E-03	9.8E-04	1.6E-03
Operational Monitoring Fourth Quarter 2001						
^{241}Am	<MDC	NA	4.7E-08	<MDC	1.2E-04	4.8E-04
^{238}Pu	<MDC	NA	6.1E-08	<MDC	1.6E-04	6.2E-04
$^{239,240}Pu$	<MDC	NA	5.5E-08	<MDC	1.5E-04	5.6E-04
^{241}Pu	<MDC	NA	2.1E-05	<MDC	5.3E-02	2.1E-01
^{228}Th	3.9E-07	5.0E-08	9.7E-08	3.9E-03	5.1E-04	9.9E-04
^{230}Th	2.2E-07	4.6E-08	1.0E-07	2.2E-03	4.7E-04	1.0E-03
^{232}Th	1.7E-07	2.7E-08	4.8E-08	1.7E-03	2.8E-04	4.9E-04
^{234}U	2.8E-07	4.6E-08	7.2E-08	2.9E-03	4.7E-04	7.4E-04
^{235}U	<MDC	NA	1.0E-07	<MDC	3.3E-04	1.1E-03
^{238}U	1.9E-07	4.1E-08	9.7E-08	1.9E-03	4.2E-04	9.9E-04

Table continued on next page

Table 12. Results of Actinide Analyses for Quarterly FAS Composite Samples (cont.)

Radionuclide	Activity Concentration ($Bq\ m^{-3}$)			Activity Density ($Bq\ g^{-1}$)		
	[a]C	[b]SD	[c]MDC	C	SD	MDC
Operational Monitoring First Quarter 2002						
^{241}Am	<MDC	NA	6.0E-08	<MDC	NA	2.7E-04
^{238}Pu	<MDC	NA	7.3E-08	<MDC	NA	3.3E-04
$^{239,240}Pu$	<MDC	NA	7.5E-08	<MDC	NA	3.5E-04
^{241}Pu	<MDC	NA	1.9E-05	<MDC	NA	8.7E-02
^{228}Th	4.3E-07	5.8E-08	1.0E-07	2.0E-03	2.7E-04	4.7E-04
^{230}Th	5.3E-07	7.5E-08	1.4E-07	2.4E-03	3.4E-04	6.6E-04
^{232}Th	3.5E-07	4.7E-08	5.8E-08	1.6E-03	2.2E-04	2.6E-04
^{234}U	6.7E-07	5.9E-08	6.4E-08	3.1E-03	2.7E-04	3.0E-04
^{235}U	<MDC	NA	6.1E-08	<MDC	NA	2.8E-04
^{238}U	6.7E-07	5.8E-08	4.4E-08	3.1E-03	2.7E-04	2.0E-04
Operational Monitoring Second Quarter 2002						
^{241}Am	<MDC	NA	5.13E-08	<MDC	NA	4.6E-04
^{238}Pu	<MDC	NA	7.22E-08	<MDC	NA	6.5E-04
$^{239,240}Pu$	<MDC	NA	5.87E-08	<MDC	NA	5.3E-04
^{241}Pu	<MDC	NA	1.58E-05	<MDC	NA	1.4E-01
^{228}Th	4.39E-07	5.86E-08	1.09E-07	3.9E-03	5.3E-04	9.8E-04
^{230}Th	3.88E-07	6.03E-08	1.14E-07	3.5E-03	5.4E-04	1.0E-03
^{232}Th	3.86E-07	4.81E-08	6.42E-08	3.5E-03	4.3E-04	5.8E-04
^{234}U	3.74E-07	4.73E-08	7.44E-08	3.4E-03	4.2E-04	6.7E-04
^{235}U	<MDC	NA	6.16E-08	<MDC	NA	5.5E-04
^{238}U	4.51E-07	5.06E-08	5.35E-08	4.0E-03	4.5E-04	4.8E-04
Operational Monitoring Third Quarter 2002						
^{241}Am	<MDC	NA	6.08E-08	<MDC	NA	3.5E-04
^{238}Pu	<MDC	NA	3.45E-08	<MDC	NA	2.0E-04
$^{239,240}Pu$	<MDC	NA	4.95E-08	<MDC	NA	2.9E-04
^{241}Pu	<MDC	NA	1.84E-05	<MDC	NA	1.1E-01
^{228}Th	3.71E-07	5.77E-08	1.22E-07	2.2E-03	3.4E-04	7.1E-04
^{230}Th	3.59E-07	6.07E-08	1.21E-07	2.1E-03	3.5E-04	7.0E-04
^{232}Th	2.40E-07	3.98E-08	7.18E-08	1.4E-03	2.3E-04	4.2E-04
^{234}U	3.37E-07	4.48E-08	6.57E-08	2.0E-03	2.6E-04	3.8E-04
^{235}U	<MDC	NA	6.60E-08	<MDC	NA	3.8E-04
^{238}U	3.30E-07	4.38E-08	6.36E-08	1.9E-03	2.5E-04	3.7E-04

[a]C = concentration
[b]SD = 1 standard deviation
[c]MDC = minimum detectable concentration
[d]NA = not applicable
[e] ^{241}Pu = ^{241}Pu analyses were not performed on the samples until January 2001
[f]NR = data not reported

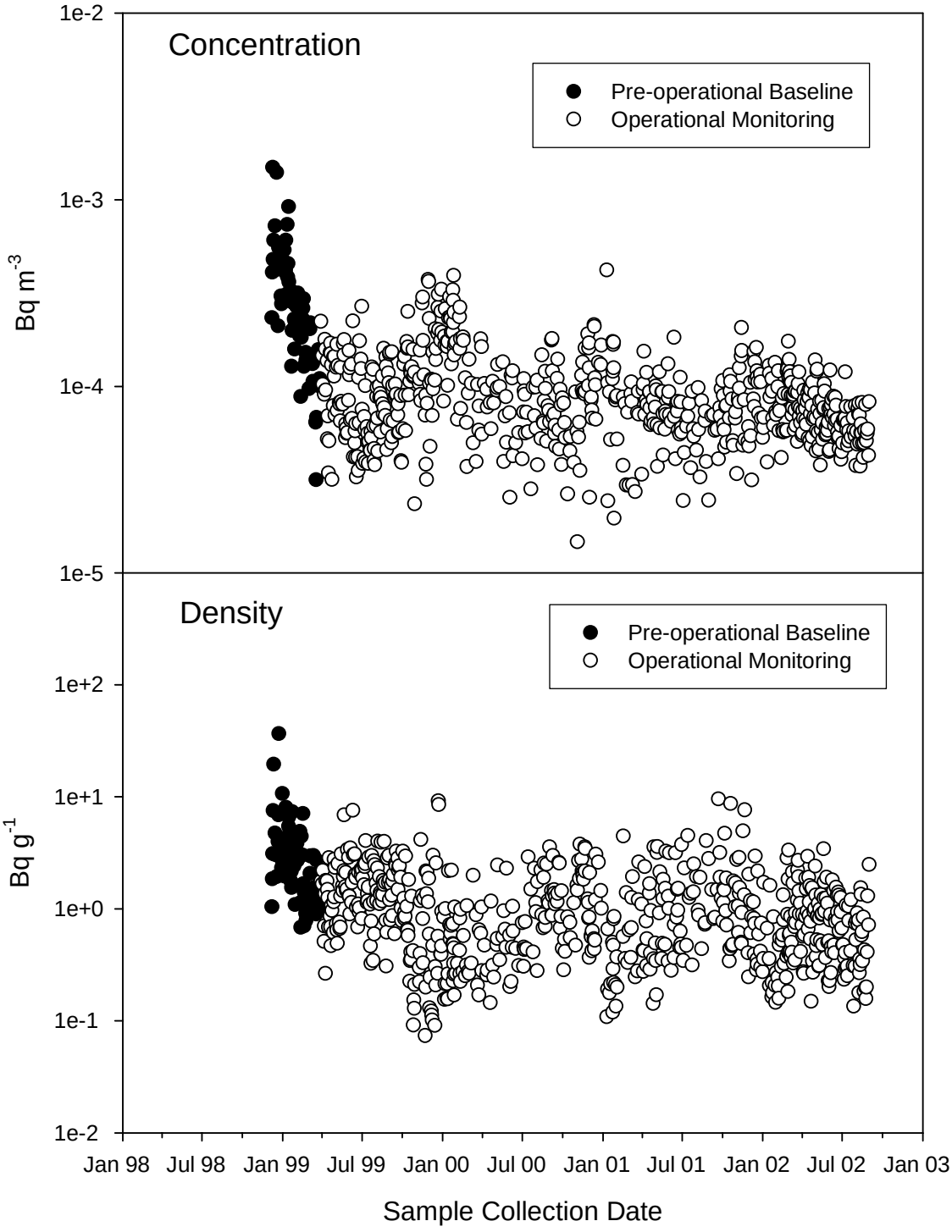

Figure 14. Alpha Emitting Radioactivity (>MDC) in FAS Samples Collected during December 1998 - September 2002

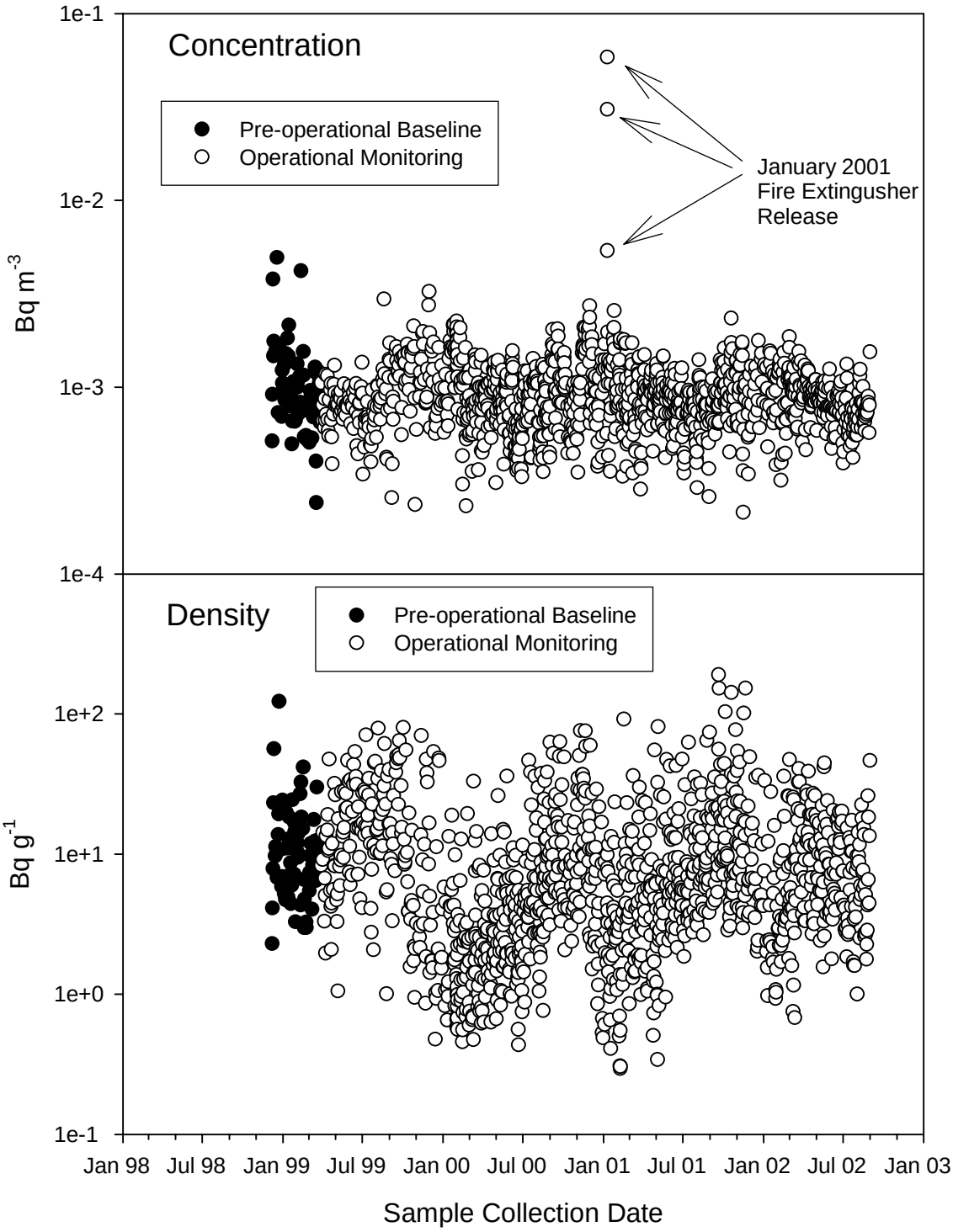

Figure 15. Beta Emitting Radioactivity (>MDC) in FAS Samples Collected during December 1998 - September 2002

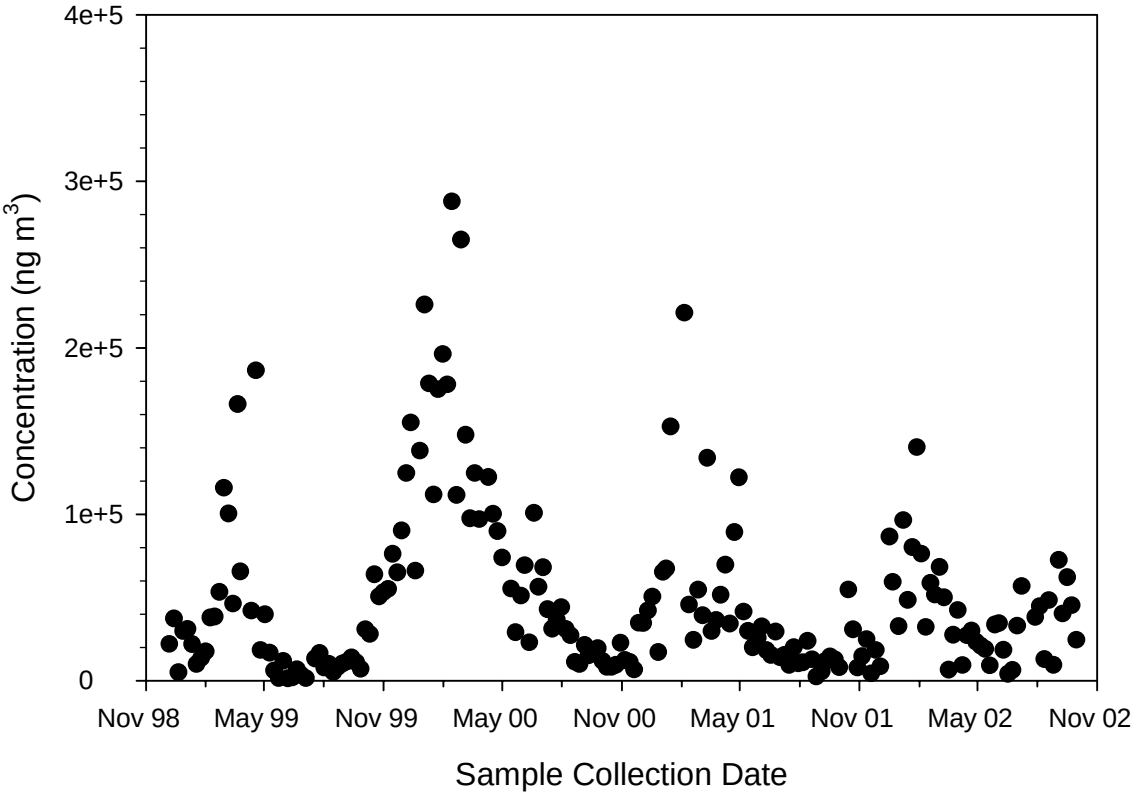

Figure 16. Sodium Released as Aerosols in the Exhaust from the WIPP

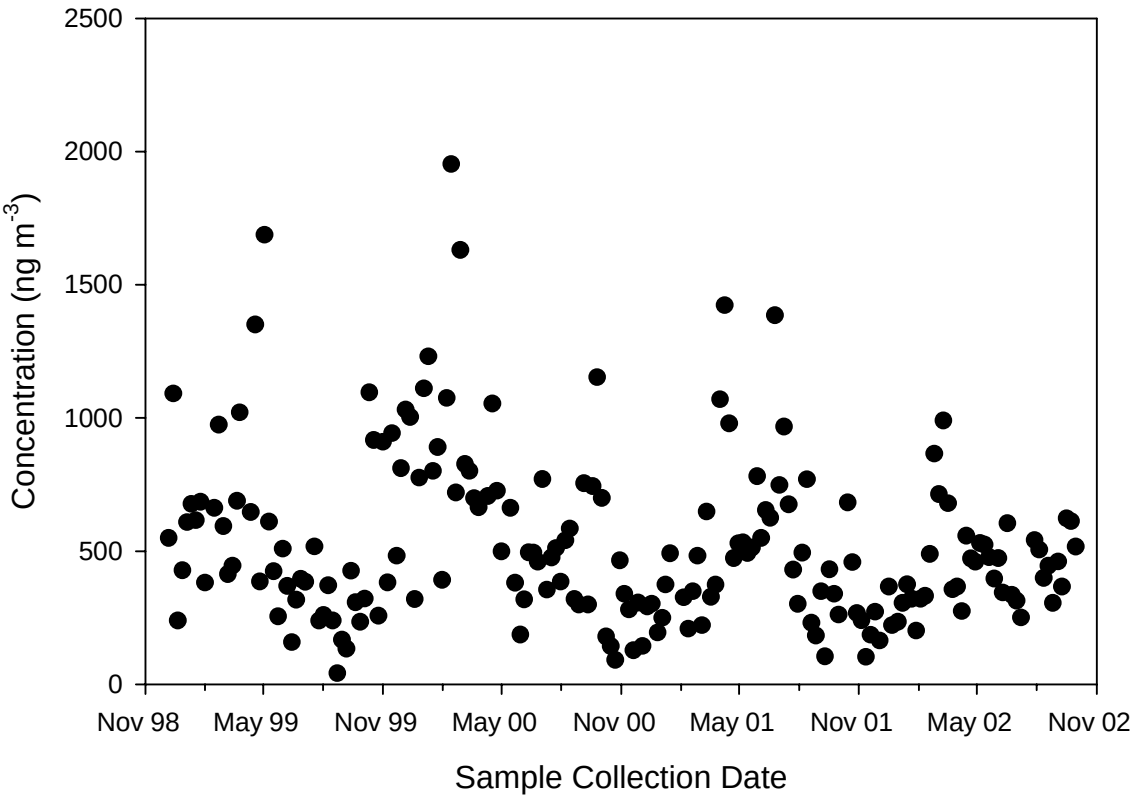

Figure 17. Aluminum Released as Aerosols in the Exhaust from the WIPP

Vertical Distribution of ^{137}Cs in the Vicinity of the Waste Isolation Pilot Plant

The distribution of ^{137}Cs activity with depth in soil profiles is utilized in this study to evaluate the importance of transport mechanisms affecting the fate of radionuclides in the arid environment of the northern extremity of the Chihuahuan desert where the WIPP site is located (Fig. 18). Fallout from aboveground nuclear testing was the primary source of ^{137} Cs in soils (Ritchie, 1990), although a potential source in the near vicinity of the WIPP site is the Gnome test site, about 8.8 km southwest of the WIPP site. This test involved a 3.1 kiloton yield nuclear underground detonation in 1961 from which venting to the atmosphere occurred (Faller, 1994).

Kirchner et al. (2002) utilized surface soil data (2-cm depth) collected using a grid scheme to characterize radionuclide concentrations in soils near the WIPP. The activity concentrations of ^{239,240}Pu, excluding one value that was less than the minimum detection level (MDL), ranged from 0.015 to 0.51 Bq kg^{-1}. The ^{137}Cs activity concentrations for the surface soils, also excluding one value <MDL, ranged from 0.31 to 15 Bq kg^{-1}, a range of more than an order of magnitude. The excluded values were associated with the same sample. The mean ^{137}Cs activity concentration from soils collected in the near vicinity of the WIPP was 3.1 (S.E. = 0.23) Bq kg^{-1} and was slightly higher for soils at a reference site approximately 20 km SE of the WIPP site, where mean ^{137}Cs activity concentration was 6.2 (S.E. = 0.52) Bq kg^{-1}. Variability in surface soil concentrations reported in this study was attributed to redistribution of contaminated soil particles via resuspension and erosion, and perhaps from variation in rates of vertical transport.

The variability in ^{137}Cs activity concentrations reported by Kirchner et al. (2002) in surface soil points to the dynamic complexity of the soil landscape. The landscape surrounding WIPP is dominated by sandy soils on undulating plains and low hills. The soils developed in noncalcareous, wind-worked sandy deposits are highly susceptible to wind and water erosion (Chugg et al., 1971). Understanding the relative role of redistribution of soil within a system, and how that redistribution depends on episodic events, is directly relevant to addressing the general issue of contaminant transport in semiarid ecosystems (Whicker et al., 2002).

The objective of this investigation was to evaluate whether some of the variability in surface concentrations of radionuclides could be explained by spatial variability in vertical transport and soil erosion processes, and whether a sufficient change in surface concentrations due to erosion could occur as to be misinterpreted as an indication of a release of contaminants from the WIPP site.

^{239,240}Pu is the radionuclide of most concern in terms of a potential release from the WIPP. ^{239,240}Pu is generally considered to have low mobility in the environment because it tends to have low solubility. However, the vertical distribution of ^{137}Cs was investigated rather than the distribution of ^{239,240}Pu because the activity concentrations of ^{137}Cs in soils from fallout are considerably greater than those for ^{239,240}Pu. In addition, ^{137}Cs is a gamma-emitter and it requires little effort to prepare samples for analysis. Although chemically quite different, ^{137}Cs is expected to behave similarly to ^{239,240}Pu in soils because it adheres tightly to cation-exchange surfaces and is relatively inert chemically (Coppinger et al., 1991).

^{137}Cs from fallout has frequently been used to characterize soil particle transport (Schimmack et al., 2002; Pimentel et al., 1995). Cesium can adsorb to organic matter as well as soil mineral particles. However, cesium adsorption on clay minerals is strongly specific, whereas adsorption on organic matter is considered to be non-specific (Staunton and Levacic, 1999). Therefore, even when organic matter is responsible for a large proportion of soil cation exchange capacity trace amounts of radiocesium will be preferentially adsorbed on clay minerals. Nevertheless, organic matter can play a role in the mobility of cesium in the soil. There is increasing evidence that

bioavailability increases with increasing organic matter content.

Given that ^{137}Cs binds preferentially to soil mineral particles, and assuming that deposition onto a landscape was uniform, then the current distribution of ^{137}Cs reflects integrated effects of water, wind, soils, vegetation, and topography on erosion and deposition during the 35 to 45 years after the major deposition period (Walling and Quine, 1991). The horizontal movement of ^{137}Cs in soils by biological and chemical processes is small in comparison with movement of ^{137}Cs by physical processes (water and wind) that transport soil particles across the landscape (Ritchie and McHenry, 1990). Coppinger et. al. (1991) examined patterns and assessed transport mechanisms of soil erosion and deposition in a native sagebrush steppe landscape in south central Wyoming using ^{137}Cs. Results showed that wind was the major factor responsible for small-scale (0.5-10 m) transport of soils in this landscape and that there was little or no intermediate-scale (approximately 100 m) transport occurring.

Downward migration of Cs is dependent on numerous factors: sorption and desorption processes in soil, soil mineral composition, soil moisture and precipitation, particle size and specific surface area, soil type, pH, organic matter content, cation competitive effects, etc. (Barisic et al., 1999). Increased concentrations of competing ions such as Na, K and H slightly decrease ^{137}Cs adsorption (Ritchie and McHenry, 1990). Adsorption of ^{137}Cs on soil particles is considered to be rapid, yielding a distribution in undisturbed soil profiles that shows an approximately exponential decrease with soil depth (Ritchie and McHenry 1990; Barasic et. al., 1999). Isaksson et. al. (2001) examined the depth distribution of ^{137}Cs in labeled soils annually over a six-year period in southern Sweden. They found great similarities between years, indicating that once deposited, ^{137}Cs migrated into the topsoil, and thereafter showed a slow migration.

This study examines the vertical distribution of ^{137}Cs and the characterization of four soil profiles in the vicinity of the WIPP site. These distributions can provide insight into the dynamics of cesium transport over the last four decades in a desert environment. In addition, results of an experiment are reported that was conducted to estimate the rate of vertical transport of cesium in WIPP soils due to the infiltration of water. In this experiment, a thin layer of soil spiked with ^{134}Cs was applied to the surface of eight columns of soil and leached with a volume of artificial rainfall approximating 30 years of rainfall. The soil was typical of the sands that dominate the WIPP soil environment. This experiment helps to evaluate whether vertical transport via de-sorption, diffusion or flow, and re-adsorption is likely to be responsible for the observed depth distributions of ^{137}Cs.

Methods

Field site profiles

Four soil profiles were sampled in the spring of 2001 for the purpose of defining the distribution of ^{137}Cs, as a function of depth. Individual profiles (Fig. 19) were sampled using the depth increments listed in Table 13. When an indurated layer was encountered at a depth shallower than 150 cm, sampling was terminated within the last unconsolidated layer at the assigned sampling thickness.

Sampling site locations were determined by the nature of the surface (minimal disturbance, absence of brush vegetation, etc.) and within a 50-m square centered on a designated grid node marker. Two sites were located within a grid surrounding the WIPP site and two sites were located within a reference grid approximately 20 km SE of the WIPP site (Kirchner et. al., 2002), (Fig. 18).

All sampling equipment (buckets, scoops, etc.) was non-metallic with the exception of excavation equipment (shovels, chipping bars, hammers). The vertical profile face was scraped clean with a plastic trowel after excavation. Samples were collected by depth increment by removing a 0.25m^2 area beginning at the profile face and working back into the undisturbed profile (Fig. 20). A 25-cm alley was removed from around the sampled area to the depth sampled before the next depth increment was collected. Soil samples were sealed in plastic containers for transport to the lab.

Soil samples were air-dried in the laboratory and sieved through a 2-mm sieve. Particle-size distribution was determined using the pipette method described by Gee and Bauder (Gee and Bauder, 1986). Specific conductivity and pH were determined on 1:2 (soil:deionized water) suspensions. Activity concentrations of ^{137}Cs in the soil samples were measured with high purity Ge gamma-spectroscopy.

Column experiment

Eight soil aliquots were prepared by spiking them with ^{134}Cs. The ^{134}Cs activity in the spiked aliquots ranged from 1310 to 1380 Bq. The spiked aliquots were subjected to three wetting and air-drying cycles with stirring performed at least five times per day. Adsorption of the ^{134}Cs to the soil particles was verified by extracting an aliquot of the water used in the last wetting cycle, filtering it and determining its ^{134}Cs activity. Three wetting and drying cycles were determined to be sufficient to result in the absorption of all but negligible quantities of the spike.

Eight columns were prepared using soil collected from the vicinity of the WIPP site (from the soil surface to a depth of 2 cm). The soil was sieved to remove roots, leaves and rock fragments and then the columns were filled to approximately 14 cm of depth (Fig. 21). Each column consisted of a piece of PVC pipe 20 cm in diameter and approximately 22 cm long. A perforated PVC plate was attached to the bottom of the column and a fine stainless steel wire screen placed on top of the plate within the column. The columns were pretreated by leaching with 1.6 L of 0.001 mole/L $CaCl_2$ solution each day for seven days. A thin "marker layer" of white sand (washed) was added to the top of each column followed by a thin layer of soil that had been spiked with ^{134}Cs. This was overlain with a filter paper, a layer of the clean, white sand, and another filter paper that were used to minimize disturbance of the soil when water was being applied. Approximately 900 cm of water (the equivalent of about 30 years of rainfall) was then applied at a rate of 3.2 L d^{-1}.

Sampling of the columns was conducted using a special tool designed to enable thin layers to be sliced from the surface of the column. The tool consisted of a circular PVC plate cut to just fit within the cylinders. An angled slit was cut in the plate from the outer edge to near the center and a tapered cutter blade mounted below the plate along one edge of the slit. The cutter blade could be adjusted to change the width of the slit. A metal barrier on the slicing tool segregated approximately 1 cm of soil on the outer edge of the column. The outer edge was segregated because it was the portion most likely to be disproportionately contaminated by any flow down the sides of the column. The plate assembly was attached to a threaded steel rod that passed through the center of a transparent plexiglass plate that rested on the top of the soil columns. The plexiglass plate had a collar attached that ensured that the plate would be centered on the top of the column. Locking nuts on the threaded rod were used to control the depth of cut. A handle was attached to the top of the threaded rod to enable it to be turned by hand to slice a layer of soil.

In the first column sampled, the first 30 mm of soil below the white marker layer were removed in 1 mm increments. Subsequent sampling was conducted by alternating 9 mm and 1 mm layers until the bottom of the column was reached. These layers were then gamma-counted using a well detector. The results from this first set of counts indicated that the ^{134}Cs had not moved more than a few millimeters. The remaining columns were therefore sampled only to a depth of 3 cm in 1 mm slices but were preserved for additional sampling until radiometric analysis confirmed that the activity could not be detected in the deepest layers and that approximately 90% of the total activity could be accounted for in the slices and the discarded outer rim of soil.

Results

Soil Profiles

Profiles 1, 2, and 3 have similar ^{137}Cs activity distributions (Table 14, Fig. 22). The maximum activity in each of these profiles occurs in either the third or fourth cm below the soil surface ranging from 7.12 to 8.66 Bq/kg, and concentration declines approximately exponentially below the peak. The activities drop off sharply within 10 cm of

the soil surface and are at or below minimum detectable concentrations (MDC) within 20 cm below the soil surface. There is no indication that radionuclides have mobilized to any substantial degree within these profiles. It is possible that the increase in ^{137}Cs activities with a slight increase in depth could be the result of leaching. However, it is also possible that the immediate surface is characteristic of a "mixing zone" in which the sandy surface could be depositional, erosional, or both.

The radionuclide distribution in profile 4 appears to be quite different from the other three profiles (Table 14, Fig. 23). An initial interpretation of the profile might be that a pulse of ^{137}Cs activity has moved to a depth of 20 cm. However, a profile inventory of the profile would suggest otherwise. Using an estimated soil bulk density of 1.4 g/cm^3, the total ^{137}Cs activity in the first three profiles ranged from 503 (±10.0) to 797 (±8.0) Bq/m^2, two to four time higher than the profile inventory reported by Collins et al. (2001) for soils located in more equatorial latitudes. The total inventory of ^{137}Cs activity in the fourth profile is almost three times higher than the other three profiles with an activity concentration of 2,263 (±30.1) Bq/m^2 (Table 15).

Pedogenically, three of the four profiles were quite similar. Profiles 1, 2, and 3 had relatively thick sandy surfaces underlain by weakly developed subsoil horizons that are distinguished from the surface by a distinct increase in clay (loamy, mixed, thermic Arenic Ustalfic Haplargid). The clay percentages presented in Table 14 ranged from approximately 2 to 5 % in the upper 6 cm of the profiles. Clay maxima in the underlying subsoil horizons ranged from approximate 12 to 15 %. Profile 4 was unique in that the clay increase in the subsoil was absent (siliceous, thermic Typic Torripsamment). The clay content observed in Table 14 for Profile 4 reflects a uniform distribution of approximately 2 % clay, increasingly only slightly (<1 %) with depth. Because the WIPP soils are dominated by sand and because Cs has an affinity for binding to clay particles, the distribution of clay in the soil could be a factor affecting the distribution of ^{134}Cs in the soil. However, there appears to be no relationship between the percentage of clay in a soil layer and the concentration of ^{134}Cs in that layer (Fig. 23).

The salinity of these sandy soils is relatively low as illustrated by the low specific conductivity values in Table 13, given the occurrence of saline and gypsiferous outcrops in the vicinity (Chugg et al., 1971). Specific conductivity values in the profiles are generally <70 µS in the upper 40 cm of the soil profiles, with the exception of the immediate surface (upper 2 cm). The concentration of salts at the soil surface reflects the capillary rise and evaporation of moisture in the soil profile. Substantial increases in salinity occur at depths below 50 cm with specific conductivity values exceeding 100 µS, and is indicative of the effective leaching depth in these profiles. The maximum value was observed in the deepest sample collected from Profile 1, having a specific conductivity value of 190 µS. To put things into perspective, a soil is not considered saline until the specific conductivity of a saturated paste exceeds 4,000 µS (USDA, 1961).

The pH values observed in each of the profiles are uniform throughout for the most part, with pH values generally falling between 6.5 and 7.5. The majority of pH values in Profiles 2, 3, and 4, occur in the pH range of 6.5 to 7.0, while pH values in Profile 1 are dominantly between 7.0 and 7.5. The extremes observed included a low pH value of 6.26 in the second cm-increment of Profile 3 and a high pH value of 7.55 in the deepest horizon sampled in Profile 1.

The distribution of ^{137}Cs seen in profiles 1, 2 and 3 is similar to that reported by Owens et al. (1996) on field and laboratory experiments in Devon, UK. These experiments were carried out to provide empirical information on the fate of ^{137}Cs applied to different soil types over a 10-month period in order to simulate transport of fallout during individual storm events. The results suggested that most of the ^{137}Cs was contained in the top few millimeters of the soil profile, with activity concentrations declining approximately exponentially with depth. The longer-term net effect of ^{137}Cs sorption and movement within the soil profile since fallout began in the

1950's was investigated by examining contemporary soil profiles collected from stable non-eroding locations. Unlike experimentally leached columns, which represent the fate of applied radiocesium after shorter periods of time, the peak in ^{137}Cs concentration was not located at the soil surface, but a few cm below (1.5 to 3.0 cm). The location of the peak below the surface is considered to reflect the interaction of internal soil processes (such as diffusion, translocation and bioturbation) operating within the soil profile since the initiation of ^{137}Cs fallout inputs in the early 1950's, and the fact that the input of ^{137}Cs to the soil surface has declined through time since the mid-1960's.

The most likely explanation for the distribution of profile 4 is deposition and burial of the original profile after it received nuclear fallout during the 1950's and early 1960's. This is not to be unexpected in a landscape dominated by sand dunes. If the ^{137}Cs pulse is bisected at the maximum (assuming a similar profile distribution to the first three profiles), and only the underlying ^{137}Cs activities considered, the total ^{137}Cs is 855 (±17.9) Bq/m^2, which is much more in line with the other three profiles (Table 15). The presence of a buried peak in ^{137}Cs activity was also interpreted as a former surface in studies of an area in southern Zambia reported by Collins et al. (2001).

The activities of ^{137}Cs in profile 4 between the surface and a depth of 15 cm range from 2.419 to 3.309 Bq/kg. This would suggest that the source of deposition is from eroded surface soil higher in ^{137}Cs relative to deeper, subsoil activity concentrations which are generally less than 1.0 Bq/kg, but somewhat lower than the maximum concentrations observed in the near surface environments of profiles 1, 2 and 3. It would be expected that the depositional material would be a combination of material relatively high in ^{137}Cs with materials lower in activity depending on the severity of the erosion from which the depositional sand originated.

The maximum depth at which ^{137}Cs could be detected further supports the argument that the original surface of profile 4 receiving radionuclide fallout has been buried. Profiles 1, 2 and 3 had detectable concentrations of ^{137}Cs down to 20 cm. Profile 4 had detectable ^{137}Cs activity down to 40 cm. If the ^{137}Cs activity peak in profile 4 that begins at a depth of 20 cm is assumed to be the original surface, this profile would have received 20 cm of erosional sand. Accounting for this burial, the depth to which ^{137}Cs has penetrated the buried profile falls in line with the other three profiles at 20 cm.

Discontinuities in the vertical distribution of properties that affect the mobilization of ^{137}Cs, such as pH and salinity, could result in the concentration of ^{137}Cs near the discontinuity. The relative uniformity in pH and salinity in the subsurface soil does not offer an explanation for the occurrence of sub-surface maxima in ^{137}Cs concentrations. In addition, the high level of conductivity in the surface layer is indicative of vertical movement of water through evaporation and capillary flow, which would be expected to work in opposition to a downward diffusion of ^{137}Cs.

Column Study

All of the activity contained in the eight soil columns appeared to be in the top few mm of soil, i.e. there was virtually no movement of the ^{134}Cs (Table 16, Fig. 24). Only 56% of the total inventory could be accounted for in Column 1. It was postulated that the remaining inventory might be found in the white marker layer and in the soil of the outer rim, both of which were discarded in the initial sampling process of Column 1. The white marker layer, positioned between the spiked soil and the underlying soil column, was sampled in the remaining columns and all of the soil removed (soil of the outer rim, cover sand above spiked soil, filter covering spiked soil) was preserved for counting. Recovery of the ^{134}Cs spike ranged from 87.3 to 96.5% in the remaining columns. All of the remaining columns also showed a very shallow ^{134}Cs profile, similar to Column 1. It is believed that most of the remaining activity may be attached to soil that could not be removed from the filter paper that was placed directly above the labeled soil. The slight indication of ^{134}Cs movement in the columns could possibly be due to soil colloids moving out of the spiked soil or possibly leaching. Regardless

of the mechanism, the miniscule movement of ^{134}Cs into the soil columns, 7 mm, could in no way account for the ^{137}Cs activity observed in the soil profiles at depths of 30 to 40 mm.

Conclusions

In assessing both the soil profiles and soil columns, it is apparent that leaching and colloidal transport are not major factors in affecting the vertical movement of ^{137}Cs or ^{134}Cs in the soils found in the vicinity of WIPP. The lateral movement of soil by wind erosion is, by far, more responsible for the redistribution of the radionuclides in this ecosystem. The maximum ^{137}Cs activity concentrations observed in the soil profiles, ranging from 6.73 to 8.66 Bq/kg, were slightly higher than the mean values reported by Kirchner et al. (2002) of 3.1 and 6.2 Bq/kg for soil samples sampled to a depth of 2.5 cm. However, the values were well within the range of activity concentrations reported, 0.31 to 15 Bq/kg. Nevertheless, there exists the potential under erosional conditions to expose soils having higher concentrations of ^{137}Cs, and potentially higher concentrations of other radionuclides such as $^{239,240}Pu$, than the concentrations currently present in surface soils.

Soil surface concentrations of radionuclides can also affect aerosol concentrations. Arimoto et al. (2002) reported that $^{239,240}Pu$ activity concentrations in total suspended particle (TSP) samples varied strongly with season (12 to 16 nBq m^{-3}), with the highest values generally occurring in spring (March through May, a period commonly typified by frequent dust storms). The source for the plutonium was hypothesized to be from resuspended soil. The authors offered that the results of their study demonstrated that processes affecting the resuspension of $^{239/240}Pu$ and possibly other substances of concern are of great importance and must be known and quantified if any impacts of WIPP operations are to be properly evaluated. Thus the detection of increased concentrations of radionuclides in the WIPP soils or aerosols should not be automatically attributed to releases from the WIPP.

Table 13. Specific Conductivity and pH data as a function of depth for the four soil profiles

Lower depth (cm)	Specific Conductivity(µS)				pH			
	Profile 1	Profile 2	Profile 3	Profile 4	Profile 1	Profile 2	Profile 3	Profile 4
1	128	200	60	128	7.20	6.58	6.53	6.59
2	71	169	48	89	7.11	6.51	6.26	6.64
3	55	66	47	73	7.12	6.74	6.43	6.73
4	63	53	39	60	6.76	6.71	6.61	6.61
6	55	55	48	50	7.28	6.76	6.58	6.79
8	64	55	45	48	7.25	6.85	6.58	6.94
10	64	50	49	39	6.76	6.78	6.51	6.94
15	56	37	50	39	6.99	6.71	6.68	6.91
20	62	31	54	39	7.19	6.75	6.52	6.88
30	54	41	49	30	7.63	6.62	6.66	6.96
40	54	43	52	30	6.83	6.64	6.67	7.05
50	73	32	62	61	7.39	6.94	6.79	6.84
75	87	33	123	72	7.19	6.90	6.59	7.08
100	125	76		137	7.44	6.74		6.70
125	190			59	7.55			6.94
150				67				6.85

Table 14. Distribution of clay and ^{137}Cs as a function of depth in the four soil profiles

Lower Depth (cm)	Clay (%)				^{137}Cs (Bq/kg)			
	Profile 1	Profile 2	Profile 3	Profile 4	Profile 1	Profile 2	Profile 3	Profile 4
1	3.6	2.0	3.2	1.9	5.261	5.030	6.321	2.419
2	4.1	2.0	4.1	2.0	6.730	6.439	6.983	2.472
3	4.3	3.0	4.4	2.1	7.399	8.296	7.124	2.923
4	3.9	2.9	4.6	2.0	6.722	8.661	5.104	2.612
6	3.9	3.4	5.0	2.0	5.932	6.864	2.126	2.816
8	4.1	3.0	5.4	2.2	2.951	3.029	1.080	2.911
10	4.7	3.7	4.9	2.1	1.378	1.658	0.408	3.052
15	4.7	3.7	5.5	1.9	0.634	0.846	0.437	3.309
20	5.1	3.8	5.6	1.7	0.400	0.240	0.191	4.489
30	5.9	3.9	5.9	2.1	<MDC[1]	<MDC	<MDC	6.727
40	6.8	4.1	6.8	2.2	<MDC	<MDC	<MDC	2.741
50	8.3	4.2	9.0	2.3	<MDC	<MDC	<MDC	<MDC
75	9.3	5.0	12.0	2.5	<MDC	<MDC	<MDC	<MDC
100	10.4	13.0		2.8	<MDC	<MDC		<MDC
125	15.1			2.8	<MDC			<MDC
150				2.8				<MDC

[1]<MDC = less than minimum detectable concentration

Table 15. Profile inventories for ^{137}Cs

Profile	Inventory (Bq/m^2)	Variance (Bq/m^2)
1	725	7.5
2	797	8.0
3	503	10.0
4	2263	30.1
[1]4	855	17.9

[1]below the sample having the maximum concentration

Table 16. Column distribution of ^{134}Cs activity expressed as percent recovery of original spike

Column Depth (mm)	Recovered from Columns (%)							
	C1	C2	C3	C4	C5	C6	C7	C8
Total Recovery	56.4	96.3	88.6	87.3	89.6	96.5	89.6	90.5
Spiked Soil	55.8	85.8	82.8	82.0	84.1	91.3	83.6	84.7
Marker Sand	[1]ND	0.80	1.43	1.37	0.70	2.45	1.35	1.34
1	0.33	0.80	1.25	0.91	0.87	0.77	1.22	0.78
2	[2]<MDC	0.76	0.59	0.57	0.63	0.46	0.91	0.74
3	<MDC	0.16	0.27	0.21	0.51	0.25	0.37	0.41
4	<MDC	0.12	0.16	0.14	0.19	0.15	0.23	0.30
5	<MDC	0.12	0.08	0.06	0.12	0.06	0.12	0.15
6	<MDC	0.09	<MDC	0.09	0.09	<MDC	0.09	0.11
7	<MDC	0.09	<MDC	0.08	<MDC	<MDC	<MDC	0.06
8	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC
9	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC
10	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC
11	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC
12	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC	<MDC

[1]ND – Not Determined
[2]<MDC – Below Minimum Detectable Concentration

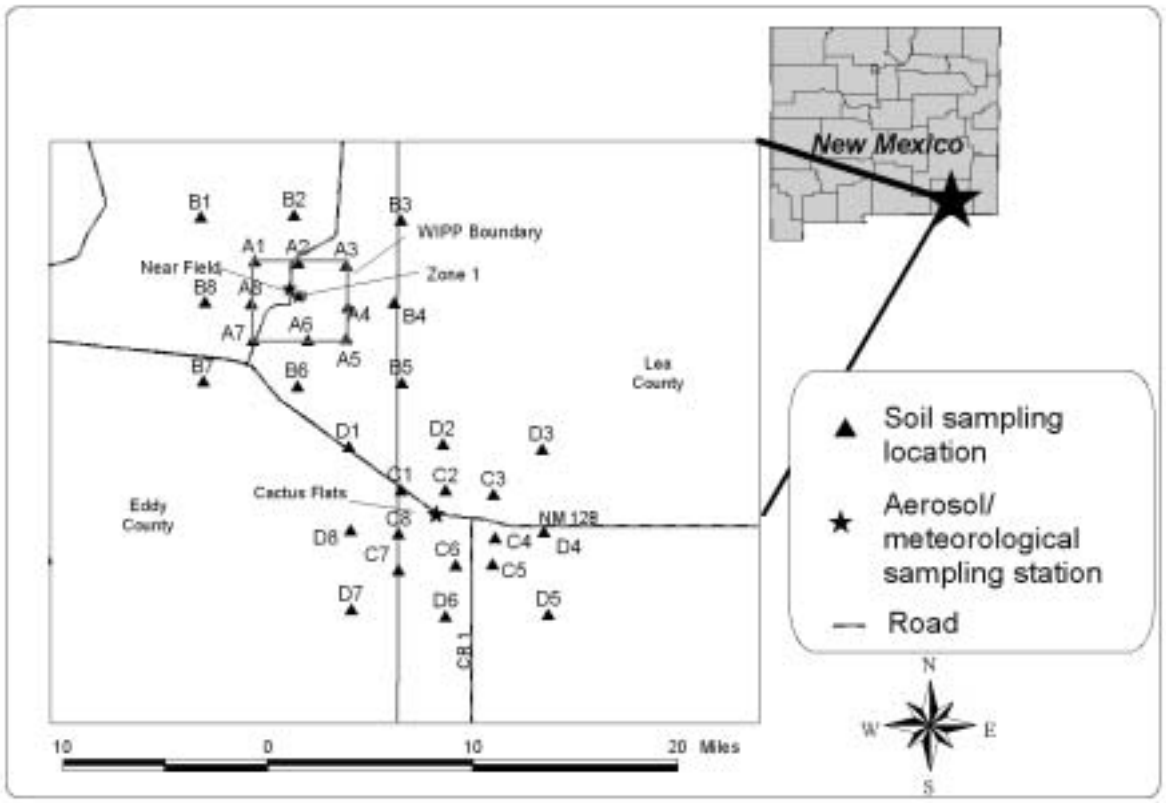

Figure 18. Site location map for profiles excavated at grid nodes A6, A8, C6, and D2

Figure 19. Soil Profile 3 before sampling

Figure 20. Soil Profile 4 after sampling

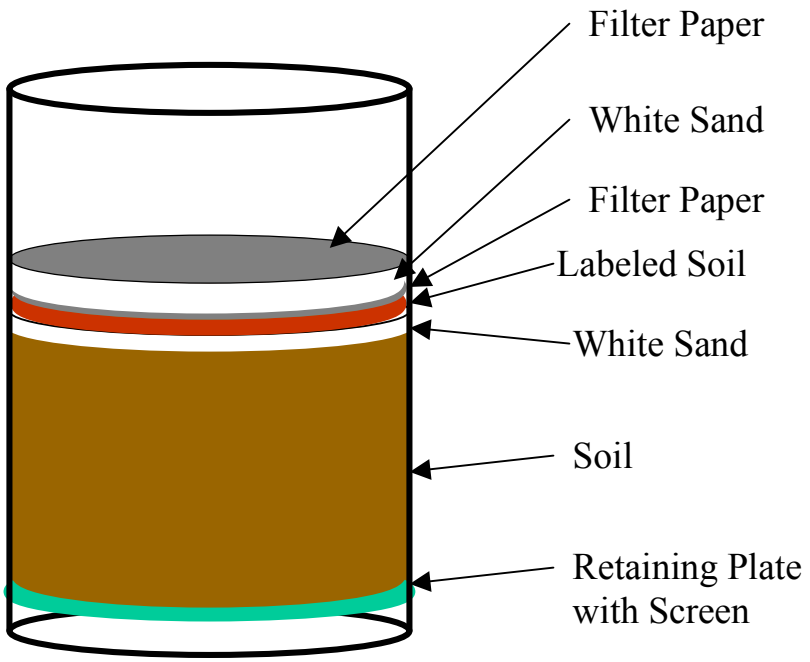

Figure 21. Schematic diagram of soil column construction

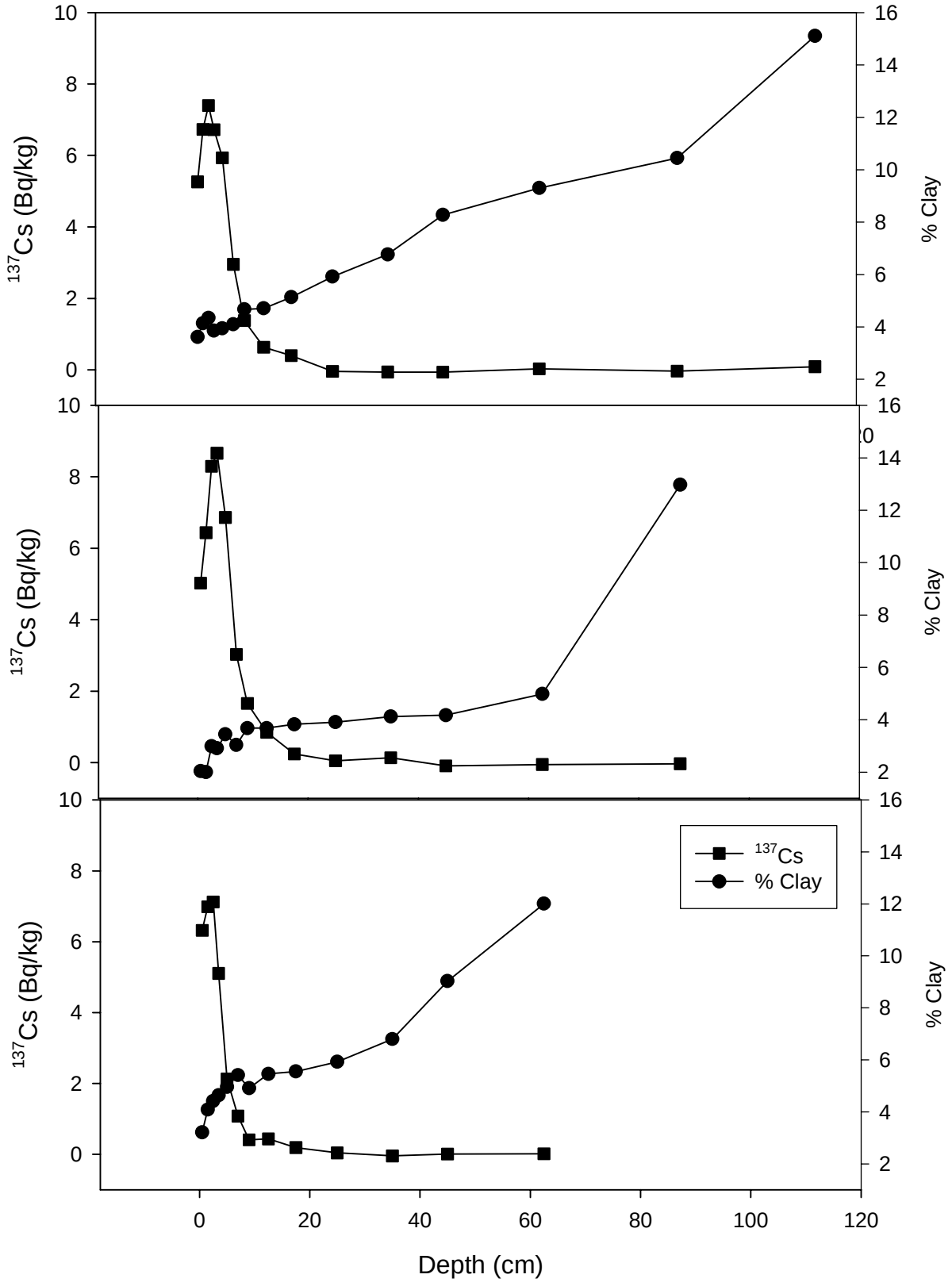

Figure 22. Distribution of ^{137}Cs and clay content expressed as a function of depth in Profiles 1, 2 and 3

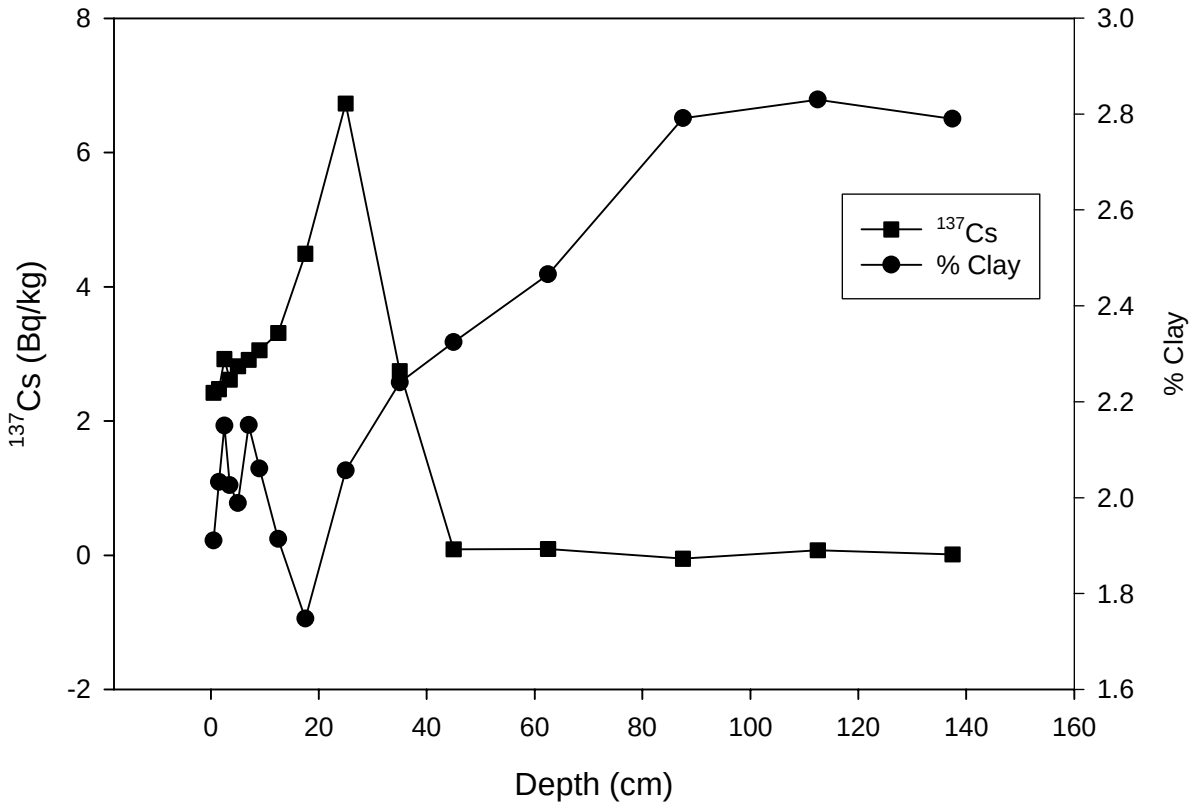

Figure 23. Distribution of ^{137}Cs and clay content expressed as a function of depth in Profile 4

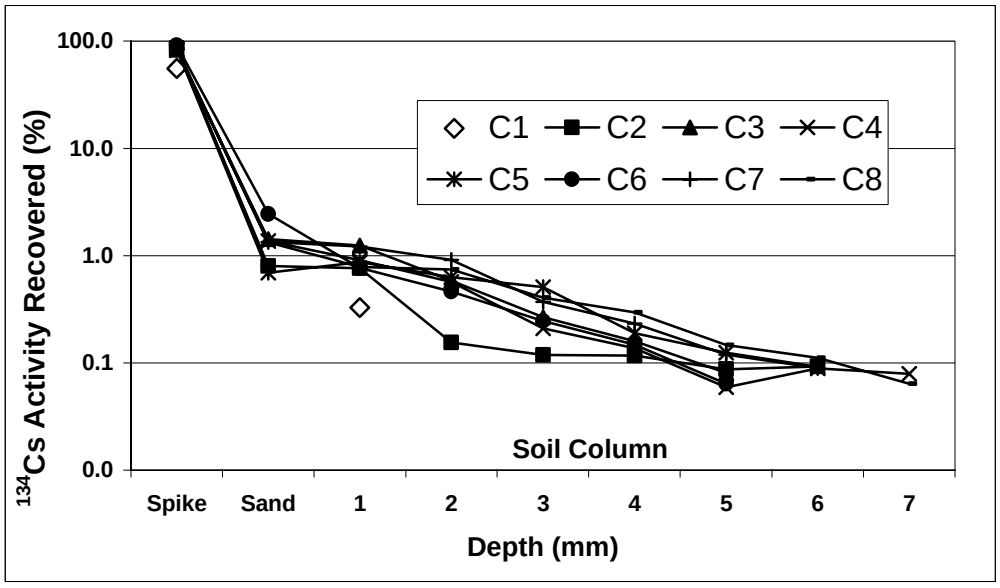

Figure 24. Distribution of ^{134}Cs activity as a function of depth in the soil columns

The Identification and Quantification of Gnome-Derived Radionuclides in WIPP Environmental Samples

Results reported herein are from soil samples collected during 2002 from nine locations near the Gnome Site. The Gnome Site lies about 9 km southwest of the WIPP boundary and was contaminated by fission radionuclides in 1961 when an underground test of a 3-kiloton ^{239}Pu device vented to the surface. This test, named Gnome, was conducted under the U.S. Atomic Energy Commission's Plowshare Program. The venting continued for about 24 h. The fallout plume was determined to extend to the northwest. Because there are elevated levels of radionuclides in the soil near the Gnome Site, there is a potential for contamination of WIPP environmental samples with soil from that location. The object of this study is to determine whether Gnome-contaminated soil can be reliably identified using isotopic ratios of plutonium, the ratios of other fission products, or ratios of non-radioactive metals. Significant differences between the isotopic and elemental ratios of Gnome-contaminated soils and those of soils collected near the WIPP site would help define a "fingerprint" or "signature" for Gnome contamination.

History of Decontamination Activities and Soil Sampling Near the Gnome Site

Decontamination of the Gnome Site was conducted in 1968-69 with the goal of removing all material exhibiting radiation levels greater that 0.1 mR h^{-1} as measured with a Geiger-Muller survey meter (Faller, 1994). The contaminated soil was either disposed of in the Gnome shaft and drift tunnels or buried on site. All surface facilities were removed and the boreholes plugged except for those used for hydrological monitoring (Faller, 1994). Starting in 1978 a second cleanup of the site was initiated. Erosion had exposed some of the contaminated material in the salvage yard and the waste dump to the northeast of the access shaft. The goal for the second remediation was to remove soil having alpha plus beta radiation exceeding 20 pCi g^{-1}.

The Gnome shaft and the nearby Coach shaft were both used for disposal of contaminated material, including a large mass of salt muck. The Coach shaft was excavated for another Plowshare program detonation that was subsequently cancelled. Materials that could not be placed in these shafts were transported to the Nevada Test Site for burial as low-level waste.

In 1992 another survey of surface activity was conducted at the Gnome Site as part of a program to assess *in-situ* gamma exposure rates for those tests conducted outside of the Nevada Test Site (Faller, 1994). Survey sites were selected based on the locations of waste disposal sites, operational facilities and shafts. Maps from previous surveillance reports were used to select sites of potential contamination and reference sites having only background levels of radiation. Twenty-two *in situ* measurements of radiation levels were collected at a 1-m height using a high purity germanium detector and a pressurized ion chamber. In addition, 11 soil cores were collected. The soil cores underwent gamma detection and 2 of the samples underwent chemical separation of ^{90}Sr for beta counting. Dose rates for ^{40}K, ^{232}Th, ^{238}U, ^{7}Be, and ^{137}Cs were reported. No determinations of actinide levels were performed. The greatest level of ^{137}Cs was measured at the site of the decontamination pad and was reported to be about 10-kBq-m^{-2}. The ^{90}Sr/^{137}Cs ratio was reported to be about 0.03, which was consistent with such measurements made during the 1978 cleanup operations. It was noted that this ratio was considerably smaller than the ratio found in the water taken from a U. S. Geological Survey well (Faller, 1994). This well was part of a tracer study conducted by the USGS that injected ^{90}Sr and ^{137}Cs into the Culebra Dolomite aquifer.

The EEG and its subcontractor, the Chemrad Tennessee Corporation (CTC) conducted another survey of the area around the Gnome Site over a seven-month interval in 1994-1995 (Kenney et al., 1995). Gamma surveys were conducted by EEG using a

sodium iodide detector positioned at 10 cm above the surface. CTC made measurements of beta, gamma and dose rates. In addition, soil samples to a depth of 2.5 cm were collected for gamma analysis using a Canberra high purity germanium detector. Aliquots of the soil underwent separation and purification in preparation for alpha spectroscopy. The four samples reported for the Gnome area sampling ranged from 0.6 mBq g^{-1} to 48,000 mBq $g^{-1}$of ^{239,240}Pu. Of 9 soil samples collected from around the WIPP site at the same time, six ^{239,240}Pu values reported as 0 (i.e. < minimum detection levels) while the remaining three ^{239,240}Pu values ranged from 0.37mBq g^{-1} to 0.74 mBq g^{-1}. Thus, in spite of the earlier remediation efforts at the Gnome Site, there are some areas where levels of radionuclide contamination remain relatively high.

Methods

Sample collection

IT Corporation conducted the most recent survey of the Gnome Site for the Nevada Operations Office starting in February 2002. IT cooperated with CEMRC by collecting Gnome Site soils for our analyses. Soil was collected using IT's standard methodology at three locations: an area along the road to the dump site, near the vent, and along the path of the plume. Three samples of approximately 1.5 kg each were collected at each location to a depth of about 2.5 cm using a plastic scoop and stainless steel containers. The soil samples were assigned Sample Identification Numbers using the SID Database system. Locations of the samples were identified using GPS and recorded. Chain of custody forms were completed on site and turned over to CEMRC with the samples. Preliminary screening showed that there are elevated levels of radioactivity in all of the samples as compared to the WIPP baseline data, but no samples showed levels where radiation safety would be a significant issue.

Measurement of Radionuclides

Initial preparation of the samples for radiological analyses consisted of passing the soil through a 2-mm sieve to remove rocks, roots and other materials. Approximately 300-mL (500-g) aliquots were used for gamma spectroscopy analysis which was conducted using high purity Ge (HPGe) detector systems for 1-2 days. A set of soil matrix standards was prepared using NIST traceable solutions and used to establish matrix-specific calibration and counting efficiencies.

A 2-g aliquot was used for analyses of each of the soil samples for ^{234}U, ^{235}U, ^{238}U, ^{230}Th, ^{232}Th, and ^{228}Th. A 10-g aliquot was used for analysis of ^{241}Am and ^{239,240}Pu for eight of the nine soil samples. The remaining sample was determined to have elevated ^{241}Am levels, hence it was deemed likely to have an elevated ^{239,240}Pu concentration. Three 1-g aliquots of this sample were analyzed for ^{241}Am and ^{239,240}Pu. The aliquots were taken from the 500-g sample used for gamma spectroscopy analysis. The aliquots were heated in a muffle furnace at 500 °C to combust organic material and spiked with a radioactive tracer to allow determination of the efficiency of extraction. The aliquots used to determine ^{241}Am and ^{239,240}Pu then underwent dissolution with HNO_3, HF and HCl followed by digestion with perchloric acid ($HClO_4$) to remove silica. These samples were then dissolved in dilute HNO_3 and H_3BO_3. The aliquots used to determine U and Th were spiked with radioactive tracers and then underwent NaOH fusion. The fused sample was dissolved in HCl and treated with HF and perchloric acid to remove silica.

Multiple precipitation, co-precipitation and ion-exchange and/or extraction chromatography procedures were then used to separate and purify the desired elements from each of aliquots. The elements of interest from each were then co-precipitated with NdF_3, deposited onto filters, mounted and counted on an alpha spectroscopy system.

Measurement of Inorganic Analytes

Soil sample aliquots of 0.25 g were analyzed for selected inorganic analytes. Aliquots were extracted from the 500 g aliquots prepared for gamma analysis. In

addition, three additional aliquots were taken from the un-sieved samples and analyzed to determine whether sieving impacts the concentration of metals in the soils. A duplicate of one of the three un-sieved soil aliquots was also analyzed. EPA Method 3052 (microwave digestion) was used to prepare samples for ICP-MS. Concentrations of Ag, Al, As, Ba, Be, Ca, Cd, Ce, Co, Cu, Dy, Er, Eu, Fe, Ga, Gd, Hg, K, La, Li, Mg, Mn, Mo, Na, Nd, Ni, Pb, Pr, Sb, Sc, Si, Sm, Sr, Th, Ti, Tl, U, V and Zn were determined using EPA Method 200.8. Levels of Cr could not be reported due to an analytical problem. The lower detection limits are in the low parts per billion range. A summary of QA/QC methods for inorganic analyses is presented in the Quality Assurance section of the Overview. The mean concentrations of these analytes reported herein for soils include only those values that are above detection levels. Thus, some estimates of the mean may be biased toward larger values.

Reported concentrations are blank-corrected. Negative concentrations of analytes can result when both the sample and the blank have concentrations above the MDL, and are hence regarded as detectable quantities, with the blank concentration exceeding the sample concentration. Thus negative values are invariably small and represent values very near the blank concentrations.

Determination of ^{239}Pu:^{240}Pu and ^{241}Am:^{239,240}Pu Ratios

The isotopes ^{240}Pu and ^{241}Pu are produced through neutron capture by ^{239}Pu and ^{238}U. ^{241}Pu subsequently decays to ^{241}Am with a half life of 13.2 years. Isotopic ^{240}Pu:^{239}Pu ratios have been used to distinguish between fallout from low yield events, such as the tests conducted at the Nevada Test Site (NTS), and global fallout, which is derived primarily from high-yield thermonuclear devices (Krey et al., 1990). ^{240}Pu:^{239}Pu atom ratios in fallout from thermonuclear devices tended to be larger than the NTS ratios due to the larger neutron fluxes prevalent in thermonuclear events (Krey et. al., 1990). For example, the atom-ratio for the MIKE shot of October, 1952 was 0.35 (Perkins and Thomas, 1980). These large thermonuclear devices were capable of injecting radioactivity into the stratosphere where it circled the globe and became the primary source of global fallout. The smaller yield tests conducted at the NTS typically injected radioactivity only into the troposphere and produced fallout largely within the southwestern United States. The NTS attempted to conduct tests only when the winds would carry fallout north over largely unpopulated areas, although some fallout plumes did extend to New Mexico.

The Gnome test employed a low yield device. Thus, Gnome fallout is expected to have a lower ^{240}Pu:^{239}Pu ratio than that of global fallout. In addition, the ^{241}Am:^{239,240}Pu ratio is expected to be elevated in global fallout compared to fallout from Gnome because ^{241}Pu is produced by the same mechanism as ^{240}Pu. ^{241}Am:^{239,240}Pu ratios can be determined by alpha spectrometry. However, ^{240}Pu:^{239}Pu ratios can not be determined by alpha spectrometry and must instead be measured using mass spectrometry (McAninch et al., 2000).

Dr. Terry Hamilton of Lawrence Livermore National Laboratory (LLNL) conducted a preliminary analysis to help evaluate the detection level for the ^{240}Pu:^{239}Pu ratio in Gnome soil using the LLNL AMS. An aliquot of one of the nine samples of soil collected from the vicinity of the Gnome site was prepared for analysis at CEMRC. The sample selected had the highest gamma activity of the nine samples (Sample Location 5 in Table 17). A 50-g aliquot of the sample was leached using acid and sub-samples of the leachate, representing 1%, 5% and 10% of the leachate by mass, were prepared in order to evaluate the detection level of the AMS methodology.

Results and Discussion

Effects of Sieving

Soils are sieved as part of the sample preparation for gamma analyses in order to remove twigs, leaves and other non-soil objects. However, sieving has the potential for contamination of the soil with metals from the sieve. The results of the inorganic analyses for the sieved and un-sieved soils

(Fig. 25) were paired by sample and relative percent differences (RPDs) by analyte were computed (Table 18). In computing RPDs the absolute value of the difference is usually used because the sign of the difference is usually not important. However, in computing the RPDs for this comparison the absolute value was not taken because the sign can be used to determine whether the concentration of a metal was higher or lower in the sieved versus un-sieved aliquot. Positive RPDs result when the sieved soil had a greater concentration of an analyte than an un-sieved soil. These results do not show any obvious impacts of screening on the metal concentrations.

Concentrations of Inorganic Analytes

The mean concentrations of most inorganic analytes in the Gnome soils were elevated compared to the mean concentrations found in the surface soils from the Near Field and Cactus Flats sites (Table 19). The data for the Cactus Flats and Near Field sites, referred to collectively as the WIPP data, are shown separately because the samples of the Cactus Flats site generally have higher concentrations of radionuclides and non-radioactive metals than does the Near Field site. The greatest differences are for Ca and Hg. Excluding these two analytes the average of the ratios across analytes shows the Gnome samples to have about three times the concentration of the metals than found in the WIPP surface soil. As discussed elsewhere (Kirchner et. al., 2002), soil texture can have a significant impact on the concentrations of radionuclides and metals. In general, there is a positive correlation between the proportion of fine particles and the concentration of radionuclides, Al and other metals. Normalizing the Near Field, Cactus Flats and Gnome results by dividing by their respective mean concentration of Al helps correct for the potential effect of soil texture differences (Table 20). The RPDs of most of these normalized analyte values are similar in magnitude to the RPDs from duplicate analyses within a location. However, the levels of Hg, Ca and Pb appear to be elevated in the Gnome samples.

The cleanup activities at the site could be responsible for elevated Pb concentrations in the soils. In any case Pb is likely to be too common in other sources of contamination to be used as a reliable fingerprint of Gnome contamination.

The higher concentration of Hg is at least partially an artifact of using a method for its analysis that has a higher MDC (average MDC = 0.017) than the method used for many of the WIPP samples (average MDC = 0.0027). The concentrations of Hg in the majority of the WIPP samples were considerably lower than the Gnome MDC and thus produce a considerably lower mean Hg concentration.

The concentration of Ca is undoubtedly a real phenomenon but one that is likely to be the result of disturbance of the soil rather than contamination of the soil from the fallout plume. Calcium carbonate tends to accumulate below the surface in soils of this area and disturbance is likely to expose these accumulations.

^{240}Pu:^{239}Pu Atom Ratios

The mean atom-ratio of ^{240}Pu to ^{239}Pu across the three dilutions (1%, 5% and 10%) was 0.075±0.006. This ratio falls within the range of 1.5E-04 to .082 reported for 99 test events conducted at the NTS. Fallout in the vicinity of the WIPP is undoubtedly a mixture of global fallout from thermonuclear devices and tests conducted at the NTS. Although the ^{240}Pu:^{239}Pu atom-ratios for the WIPP soils have yet to be measured, measurements of the ratio in soils collected from around the world typically show values of about 0.19. The exceptions in the northern hemisphere are associated with soils collected in the southwestern United States, where fallout from the NTS events was also deposited (Perkins and Thomas 1980). A study of sediments from a lake in Utah can be used to estimate the expected range for the ^{240}Pu:^{239}Pu atom-ratio in WIPP soil given the potential for some contamination by NTS events. Prior to about 1960 the ^{40}Pu:^{239}Pu atom-ratios in these sediments ranged from about 0.05 to 0.10 (Krey et al., 1990). The fallout in these earlier sediments was a mixture of NTS fallout and some thermonuclear event fallout, particularly from MIKE. Thereafter the ratio increased as greater numbers of

thermonuclear devices were tested. The ^{240}Pu:^{239}Pu atom-ratios peaked in 1963 at 0.186 and then declined to about 0.14 (Krey et al., 1990). In terms of the total amount of deposition, the Krey et al. (1990) study also showed that deposition was dominated by events that occurred after 1960, i.e. during the period when ^{240}Pu:^{239}Pu atom-ratios were 0.14 or larger. The lake from which the sediments were collected lies near Salt Lake City, Utah and hence was more likely to have received fallout from the NTS than was the WIPP site. Therefore, the expected ^{240}Pu:^{239}Pu atom-ratio for the WIPP soil is expected to be larger than 0.14. Thus, the ^{240}Pu:^{239}Pu atom-ratio is likely to provide a good fingerprint of Gnome contamination, assuming that measurements of the ^{240}Pu:^{239}Pu atom-ratio confirms a ratio of 0.14 or higher in the WIPP soils.

Radionuclide Concentrations

All of the Gnome soil samples show elevated concentrations of ^{137}Cs as compared to the across-year maximum concentrations for the WIPP sites (Table 17). The maximum observed concentration of ^{137}Cs for the Gnome samples, 2.98E+03 Bq kg^{-1}, was more than 100 times larger than the largest concentration seen previously in the WIPP surface soil samples. Variability among the ^{137}Cs concentrations of the Gnome samples was high. The Gnome concentrations for ^{228}Ac and ^{40}K fell within the range of values previously measured for the WIPP soils locations except for sample 9, which shows somewhat elevated levels of both of these radionuclides. Sample 9 was collected on the path of the plume and had a noticeably greater content of clay and silt than typical for surface soils in that area. Because ^{40}K is a naturally occurring radionuclide, its elevated concentration may simply be due to differences in soil texture (Kirchner et al., 2002). Concentrations of ^{238}Pu, $^{239,240}Pu$ and ^{241}Am for one of the Gnome samples were more than 650 times greater than the maximum concentrations observed in WIPP soils. However, the remaining samples showed at most only moderately higher actinide activity concentrations and many of these samples fell within the range of concentrations previously observed in WIPP soils.

The mean ratio of ^{137}Cs to $^{239,240}Pu$ from Gnome samples is significantly greater than the mean ratio from WIPP samples ($p<0.001$) (Table 21). However, the within-sample variability is very great in these Gnome samples and there is obviously little correlation between ^{137}Cs and $^{239,240}Pu$. ^{238}Pu was above the minimum detection level in only 21 WIPP samples. The mean ratio of ^{238}Pu to $^{239,240}Pu$ from the Gnome sample is not significantly different than the mean ratio for the WIPP samples. However, the mean ratio for the WIPP samples (0.13) is dominated by two of the 21 samples. Excluding these two values reduces the WIPP mean ratio to 0.094. In either case the ^{238}Pu:$^{239,240}Pu$ ratios for both Gnome and WIPP exceed the global fallout mean ratio for latitudes of 30-40ºN of 0.061 (Mitchell et al., 1997). However, it should be noted that the mean WIPP ratio excludes all values that were less than the minimum detection level and hence the mean ratio is expected to be biased towards a value greater than the true mean.

The mean activity ratio of ^{241}Am to $^{239,240}Pu$ in the Gnome samples, 0.31, is significantly lower than that of the WIPP samples, 0.39 ($p<0.003$). The ^{241}Am is most likely the decay product of ^{241}Pu that was produced by activation during the blast. The greater neutron flux associated with the tests that contributed to world-wide fallout (Krey et al., 1990) is the probable cause for the ^{241}Am:$^{239,240}Pu$ ratio being higher in the WIPP samples than in the Gnome samples.

Conclusions

The preliminary results of AMS analysis indicate that the ^{240}Pu:^{239}Pu ratio should provide a good fingerprint for Gnome contamination and that detection levels are low enough that as little as one gram of soil at typical WIPP concentrations could provide a sufficient mass of plutonium for analysis using AMS. It should be noted that the ^{240}Pu:^{239}Pu ratio was determined for only one relatively low-activity sample, and that analyses of additional samples are required to provide confidence in the results. The plutonium in the Gnome samples is expected to be a mixture of global fallout, NTS fallout

and Gnome deposition. Thus, the sample having the highest activity concentration of ^{239,240}Pu would be of greatest value for identifying whether the ^{240}Pu:^{239}Pu atom ratio for Gnome fallout could be even lower that the 0.075 value found using the relatively low-activity sample. Although the other Gnome samples have activity concentrations much more similar to WIPP soils, the measurement of the ^{240}Pu:^{239}Pu ratio in those samples may also prove valuable for confirming a correlation with the ^{241}Am:^{239,240}Pu ratios.

The likely ^{240}Pu:^{239}Pu atom ratio for WIPP soils was only inferred from literature values for other locations. Direct measurement of WIPP ^{240}Pu:^{239}Pu ratios should be made to confirm that the average ratio is higher than that for Gnome contaminants and to help establish a range of variability on the measurements.

The ^{241}Am:^{239,240}Pu ratio could also be used as a fingerprint for the Gnome contamination. This ratio is of interest because the cost of determining this ratio using alpha spectroscopy is considerably lower than determining the ^{240}Pu:^{239}Pu ratio by AMS. It would be of value, however, to confirm that the ^{241}Am:^{239,240}Pu is correlated with ^{240}Pu:^{239}Pu ratio across the set of Gnome samples. High variability in the ^{137}Cs:^{239,240}Pu ratio suggests that this ratio would not be particularly useful for identifying Gnome contamination.

Table 17. Mean concentrations (Bq kg^{-1}) of radionuclides for each of nine Gnome sample locations and across-sample statistics in comparison to the concentrations in surface soils collected from two areas near the WIPP site.

Site	Sample Location	^{137}Cs	^{228}Ac	^{40}K	^{60}Co	^{238}Pu	$^{239,240}Pu$	^{241}Pu	^{241}Am
Gnome	1	1.98E+03	1.22E+01	3.24E+02	6.58E-01	1.05E+02	7.44E+02	7.82E+02	1.68E+02
	2	5.10E+02	1.04E+01	2.02E+02	<MDC	3.20E-02	2.42E-01	<MDC	8.21E-02
	3	4.69E+01	7.74E+00	1.77E+02	<MDC	1.36E-01	4.67E-01	<MDC	2.16E-01
	4	4.10E+02	9.10E+00	1.84E+02	<MDC	4.63E-02	1.66E-01	<MDC	6.17E-02
	5	2.98E+03	9.93E+00	1.93E+02	<MDC	3.09E-02	1.58E-01	<MDC	4.81E-02
	6	9.47E+02	9.34E+00	2.01E+02	<MDC	1.61E-02	2.90E-01	<MDC	9.26E-02
	7	1.79E+03	1.12E+01	2.38E+02	<MDC	<MDC	2.61E-01	<MDC	9.45E-02
	8	9.35E+02	9.95E+00	2.34E+02	<MDC	3.95E-02	3.90E-01	<MDC	1.01E-01
	9	3.08E+02	1.94E+01	3.40E+02	<MDC	6.56E-02	7.43E-01	<MDC	2.23E-01
Gnome	Mean	8.40E+02	1.03E+01	2.18E+02	6.58E-01	2.88E+01	1.49E+02	7.82E+02	3.61E+01
	Minimum	4.59E+01	7.62E+00	1.75E+02	6.58E-01	1.61E-02	7.28E-02	2.59E+02	4.28E-02
	Maximum	2.98E+03	1.94E+01	3.40E+02	6.58E-01	2.19E+02	1.55E+03	1.63E+03	3.46E+02
	Standard	2.47E+02	8.434E-01	1.51E+01		2.00E+01	1.05E+02	4.30E+02	2.52E+01
	n	13	13	13	1	11	15	3	14
Cactus Flats	Mean	5.63E+00	1.13E+01	2.24E+02	NA	2.27E-02	2.04E-01	NA	6.96E-02
	Minimum	6.93E-01	6.67E+00	1.42E+02	NA	1.13E-02	1.35E-02	NA	2.10E-02
	Maximum	1.48E+01	1.58E+01	3.24E+02	NA	4.23E-02	5.07E-01	NA	2.57E-01
	Standard	3.34E-01	2.31E-01	437E+00	NA	2.19E-03	1.11E-02	NA	4.16E-03
	n	86	86	86	NA	17	111	NA	86
Near Field	Mean	3.46E+00	8.56E+00	2.14E+02	NA	2.59E-02	1.19E-01	NA	4.74E-02
	Minimum	2.97E-01	5.59E+00	1.42E+02	NA	1.55E-02	1.45E-02	NA	1.29E-02
	Maximum	8.83E+00	1.37E+01	3.21E+02	NA	6.26E-02	3.89E-01	NA	1.27E-01
	Standard	2.06E-01	1.87E-01	4.18E00	NA	9.20E-03	6.80E-03	NA	2.88E-03
	n	85	86	86	NA	5	90	NA	66

Table 18. Relative percent differences between sieved and unsieved soils paired by sample

Note: Only results greater than the minimum detectable concentration are reported.

Analyte	RPD	Analyte	RPD	Analyte	RPD
Al	-8.54%	Gd	-10.35%	Sc	-2.67%
Al	-4.79%	Gd	16.09%	Sc	-3.00%
Al	32.40%	Gd	41.47%	Sc	31.61%
Al	-17.75%	Gd	-6.93%	Sc	-29.29%
As	-54.51%	K	14.42%	Si	-14.83%
Ba	-6.03%	K	11.18%	Si	-22.05%
Ba	3.96%	K	44.54%	Si	7.68%
Ba	45.66%	K	-11.45%	Si	-94.33%
Ba	-7.65%	La	-37.08%	Sm	-18.43%
Be	34.24%	La	-2.87%	Sm	9.59%
Be	28.19%	La	40.57%	Sm	42.75%
Be	57.81%	La	-15.36%	Sm	-9.71%
Be	-4.61%	Li	38.44%	Sr	-31.73%
Ca	15.01%	Li	31.59%	Sr	-28.14%
Ca	13.52%	Li	63.80%	Sr	34.02%
Ca	29.81%	Li	8.43%	Sr	-8.87%
Ca	5.04%	Mg	20.79%	Th	39.09%
Ce	-1.98%	Mg	21.43%	Th	-4.16%
Ce	-33.46%	Mg	46.70%	Th	47.91%
Ce	42.48%	Mg	4.80%	Th	3.36%
Ce	-13.17%	Mn	2.40%	Ti	-55.51%
Co	19.97%	Mn	3.34%	Ti	-50.87%
Co	25.37%	Mn	32.99%	Ti	20.31%
Co	30.60%	Mn	-5.93%	Ti	-72.89%
Co	-25.64%	Mo	-18.25%	Tl	-53.04%
Cu	-12.79%	Mo	-44.17%	U	21.58%
Dy	-1.97%	Mo	36.47%	U	-1.54%
Dy	17.27%	Mo	-59.67%	U	46.45%
Dy	34.48%	Nd	6.38%	U	-8.27%
Dy	-8.38%	Nd	-21.12%	V	-13.90%
Er	18.66%	Nd	45.49%	V	-11.64%
Er	5.09%	Nd	-6.36%	V	28.99%
Er	37.30%	Ni	16.07%	V	-29.14%
Er	-9.49%	Ni	13.78%	Zn	1.62%
Eu	-16.33%	Ni	44.64%	Zn	-0.14%
Eu	4.15%	Ni	-2.90%	Zn	24.24%

Table continued on next page

Table 18. Relative percent differences between sieved and unsieved soils paired by sample (cont.)

Note: Only results greater than the minimum detectable concentration are reported.

Analyte	RPD	Analyte	RPD	Analyte	RPD
Eu	-13.21%	Pb	8.62%	Zn	-16.24%
Fe	-4.26%	Pb	11.72%		
Fe	0.89%	Pb	31.22%		
Fe	31.72%	Pb	-4.66%		
Fe	-13.01%	Pr	3.47%		
Ga	-63.35%	Pr	-27.14%		
Ga	-64.33%	Pr	43.78%		
Ga	-21.17%	Pr	-11.16%		
Ga	-29.09%				

Table 19. Ratio of concentration of metals in sieved Gnome soils to those in WIPP soils.

Analyte	WIPP Location	Ratio Gnome/WIPP	Analyte	WIPP Location	Ratio Gnome/WIPP
Ag	Cactus Flats	6.08	Mg	Cactus Flats	3.70
Ag	Near Field	9.49	Mg	Near Field	4.28
Al	Cactus Flats	2.69	Mn	Cactus Flats	2.34
Al	Near Field	3.50	Mn	Near Field	3.09
As	Cactus Flats	1.42	Mo	Cactus Flats	2.21
As	Near Field	1.92	Mo	Near Field	3.04
Ba	Cactus Flats	4.52	Na	Cactus Flats	3.54
Ba	Near Field	6.06	Na	Near Field	3.37
Be	Cactus Flats	2.52	Nd	Cactus Flats	1.68
Be	Near Field	3.26	Nd	Near Field	2.14
Ca	Cactus Flats	27.23	Ni	Cactus Flats	2.69
Ca	Near Field	27.72	Ni	Near Field	2.86
Ce	Cactus Flats	1.99	Pb	Cactus Flats	4.43
Ce	Near Field	2.36	Pb	Near Field	6.11
Co	Cactus Flats	2.50	Pr	Cactus Flats	1.70
Co	Near Field	3.19	Pr	Near Field	2.18
Cu	Cactus Flats	6.30	Sb	Cactus Flats	3.82
Cu	Near Field	8.09	Sb	Near Field	4.51
Dy	Cactus Flats	1.99	Sc	Cactus Flats	2.64
Dy	Near Field	2.66	Sc	Near Field	3.38
Er	Cactus Flats	2.10	Si	Cactus Flats	1.96
Er	Near Field	2.84	Si	Near Field	0.78
Eu	Cactus Flats	2.62	Sm	Cactus Flats	1.86
Eu	Near Field	3.37	Sm	Near Field	2.36
Fe	Cactus Flats	1.64	Sr	Cactus Flats	5.24
Fe	Near Field	2.11	Sr	Near Field	6.49
Gd	Cactus Flats	2.01	Th	Cactus Flats	1.43
Gd	Near Field	2.61	Th	Near Field	1.84
Hg	Cactus Flats	21.39	Ti	Cactus Flats	3.82
Hg	Cactus Flats	11.51	Ti	Near Field	4.51
Hg	Near Field	25.45	Tl	Cactus Flats	3.99
Hg	Near Field	18.12	Tl	Near Field	3.87
K	Cactus Flats	2.75	U	Cactus Flats	2.16
K	Near Field	3.63	U	Near Field	2.62
La	Cactus Flats	1.72	V	Cactus Flats	3.39
La	Near Field	2.23	V	Near Field	3.62
Li	Cactus Flats	2.52			
Li	Near Field	3.00			

Table 20. Relative percent differences between the Gnome and WIPP normalized analyte concentration

Normalization involved dividing the analyte concentrations for Gnome, Cactus Flats and Near Field by their respective mean Al concentration in order to help correct for soil texture differences.

Analyte	Grid ID	RPD Of Ratios	Analyte	Grid ID	RPD Of Ratios
As	Cactus Flats	44.51%	Mg	Near Field	38.58%
As	Near Field	40.85%	Mn	Cactus Flats	8.32%
Ba	Cactus Flats	43.60%	Mn	Near Field	6.81%
Ba	Near Field	46.33%	Mo	Cactus Flats	18.86%
Be	Cactus Flats	12.96%	Mo	Near Field	24.52%
Be	Near Field	12.44%	Nd	Cactus Flats	22.55%
Ca	Cactus Flats	163.14%	Nd	Near Field	24.67%
Ca	Near Field	154.06%	Ni	Cactus Flats	19.10%
Ce	Cactus Flats	11.30%	Ni	Near Field	1.08%
Ce	Near Field	20.21%	Pb	Cactus Flats	116.46%
Co	Cactus Flats	4.33%	Pb	Near Field	120.16%
Co	Near Field	2.24%	Pr	Cactus Flats	24.30%
Cu	Cactus Flats	57.07%	Pr	Near Field	25.55%
Cu	Near Field	55.88%	Sb	Cactus Flats	73.32%
Dy	Cactus Flats	12.07%	Sb	Near Field	64.66%
Dy	Near Field	9.41%	Sc	Cactus Flats	0.63%
Er	Cactus Flats	7.72%	Sc	Near Field	0.89%
Er	Near Field	3.86%	Si	Cactus Flats	1.00%
Eu	Cactus Flats	7.48%	Si	Near Field	105.52%
Eu	Near Field	8.53%	Sm	Cactus Flats	15.64%
Fe	Cactus Flats	24.48%	Sm	Near Field	18.12%
Fe	Near Field	25.52%	Sr	Cactus Flats	55.03%
Gd	Cactus Flats	3.19%	Sr	Near Field	50.41%
Gd	Near Field	3.50%	Th	Cactus Flats	32.55%
Hg	Cactus Flats	94.38%	Th	Near Field	33.60%
Hg	Cactus Flats	135.27%	Ti	Cactus Flats	26.21%
Hg	Near Field	130.28%	Ti	Near Field	16.62%
Hg	Near Field	108.53%	Tl	Cactus Flats	10.71%
K	Cactus Flats	7.90%	Tl	Near Field	39.60%
K	Near Field	9.15%	U	Cactus Flats	2.15%
La	Cactus Flats	25.32%	U	Near Field	4.80%
La	Near Field	25.51%	V	Cactus Flats	39.69%
Li	Cactus Flats	17.54%	V	Near Field	20.60%
Li	Near Field	8.74%	Zn	Cactus Flats	17.96%
Mg	Cactus Flats	49.70%	Zn	Near Field	27.74%

Table 21. Ratios of ^{137}Cs and ^{238}Pu to $^{239,240}Pu$ in Gnome samples in comparison to mean values for WIPP soils.

Sample ID	Aliquot ID	$^{137}Cs/^{239,240}Pu$	$^{238}Pu/^{239,240}Pu$	$^{241}Am/^{239,240}Pu$
102837	102899	2.66	0.14	0.23
102838	102900	2106.22	0.13	0.34
102839	102901	55.05	0.16	0.25
102839	103029	629.97	NA	NA
102840	102902	2541.06	0.28	0.42
102840	103030	2370.86	NA	0.27
102841	102903	18853.69	0.20	0.30
102842	102904	3261.37	0.06	0.32
102843	102905	6874.66	<MDC	0.36
102844	102906	2393.92	0.10	0.26
102845	102907	414.45	0.09	0.30
Gnome	Mean	3591.26	0.14	0.31
	Standard Error	1635.70	0.025	0.02
	n	11	8	10
WIPP	Mean	29.33	0.13	0.39
	Standard Error	0.42	0.027	0.02
	n	162	21	133

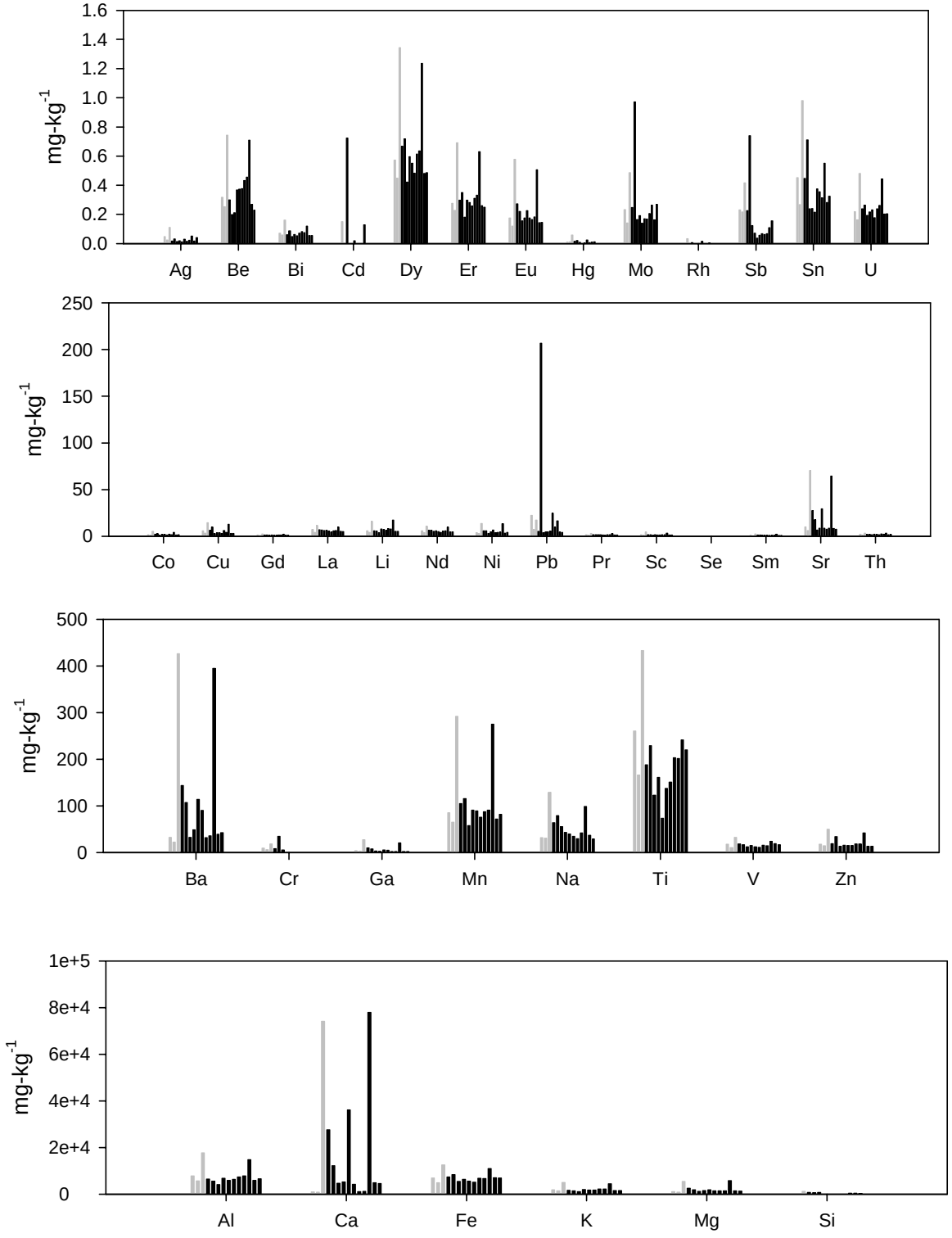

Figure 25. Concentrations of metals in Gnome soils.

Note: Screened soils are black bars, un-screened soils are gray.

Radionuclides in Oil and Gas Formations in the Vicinity of WIPP

In 1996 the National Research Council (NRC) was asked by the U.S. DOE-CBFO to identify the limiting technical components of the WIPP program, with a twofold goal of:

- improving the understanding of long-term performance of the repository and
- identifying technical options for improvements to the National Transuranic (TRU) Program without compromising safety.

One of the shortcomings identified by the NRC was an absence of radiological baseline information for subsurface brines and hydrocarbons near the WIPP site, even though there had been extensive monitoring of radioactivity in the air, soils, fluvial sediments, surface water, shallow groundwater, and populace. Therefore, the NRC recommended that the DOE develop and implement a plan to sample oil-field brines, petroleum, and solids associated with current hydrocarbon production to assess the magnitude and variability of naturally occurring radioactive material (NORM) in the vicinity of the WIPP site. The radionuclides of interest included those that contribute to NORM (^{226}Ra, ^{234}U, ^{238}U, ^{224}Ra, ^{228}Ra, ^{228}Th, ^{230}Th, ^{232}Th, ^{40}K and ^{235}U) and those present in the TRU waste inventory destined for WIPP (^{241}Am, ^{137}Cs, ^{238}Pu, ^{239,240}Pu, and ^{90}Sr). As some TRU inventory radionuclides are not commonly found in nature, it was recommended that sampling to determine whether such radionuclides are present in the environment would be an effective approach to distinguish radioactivity due to NORM from that due to TRU waste.

The report further pointed out that the issue of baseline values for NORM in the vicinity of the WIPP site is important for future monitoring of changes in radioactivity levels in and around the site. The reason for concern is that subsurface oil and gas in the vicinity of the site already contain NORM. Radioactive material in oil and gas found in the future could mistakenly be interpreted to come from the repository and thereby cast a doubt on the performance of the nearby WIPP.

In response to the NRC recommendations, in 2000 CBFO requested CEMRC undertake an investigation to determine existing levels of NORM and TRU radionuclides in the oil and gas formations in the vicinity of the WIPP site. Given the current scope of CEMRC's WIPP-EM project, monitoring of oil and gas formations is a natural extension of that work. Following CBFO's request, plans were developed and work began in earnest in 2002.

Sample Collection

The 2002 activities for the project involved sample collection, method development, and the start of the analyses for baseline characterization. It is important to point out that the WIPP began receiving TRU waste in March 1999 and TRU mixed waste in September 2000. Attempts to define background levels of radioactive elements in samples from the oil and gas formations must recognize that the characterization is being performed on samples collected after WIPP began receiving waste. However, the possibility that contamination of the oil and gas formation in the vicinity of the WIPP could have occurred from a release of radioactive elements from the WIPP is miniscule.

As a first step in designing the study a review was performed of the geologic stratigraphy of the oil and gas producing formations below the WIPP, which indicated that there are three formations that should be characterized. The Delaware Mountain group (over 385 wells in the nine quadrant region surrounding the WIPP) consists of three formations: Bell Canyon, Cherry Canyon and Brushy Canyon. Both the Cherry Canyon and Brushy Canyon formations are highly productive while Bell Canyon is not. Lying below the Delaware Group is the Bone Spring formation (approximately 54 wells in the nine quadrant region surrounding the WIPP). Below these formations are the Wolf Camp, Strawn, Atoka and Morrow formations. Most of the development in these formations has been for natural gas. The decision was made to concentrate on wells in the Delaware Group

in order to maximize the number of wells sampled.

The original project strategy was to sample oil and brine from individual pools within single stratigraphic formations. Through discussions with professionals in the field of oil well development, it became apparent that the level of detailed sampling originally sought was not feasible. A single well is developed at whatever depths in which there is potential to produce oil and therefore the representativeness of single wells is debatable. As a result, most wells available will represent a combination of oil from multiple formations occurring within the Delaware Group.

Method Development

Moreover, oil producers participating in the study recommended that sampling oil field batteries should be considered. Batteries represent inputs from a number of oil wells, usually numbering 5 to 7 wells per battery. The end result was that instead of sampling 40 individual wells, developed in multiple formations within the Delaware, 18 batteries were selected representing approximately 86 wells, yielding a substantially larger sampling population. Collection of oil and brine samples was completed in September 2002 through services provided by Perini Environmental (Carlsbad, NM). Each battery sampled by the service provider produced an individual brine and hydrocarbon sample.

Radionuclides identified for the characterization of NORM in both brine and hydrocarbon media include selected elements of the ^{238}U decay series (^{226}Ra, ^{234}U, and ^{238}U), the ^{232}Th decay series (^{224}Ra, ^{228}Ra, ^{228}Th, ^{230}Th, and ^{232}Th), as well as ^{40}K and ^{235}U. Characterization will also include selected manmade elements that are known or expected to occur in waste deposited at the WIPP (^{241}Am, ^{137}Cs, ^{238}Pu, ^{239,240}Pu, and ^{90}Sr).

Although NORM radioactivity is generally concentrated in the water fraction of brine/hydrocarbon mixtures, research investigations characterizing this phase preference for the targeted radionuclides found in TRU waste could not be found. This will necessitate the eventual characterization of the hydrocarbon phase as well. Method development activities in 2002 concentrated on the brine phase only.

A flowchart is presented in Fig. 26 that illustrates the analyte separation strategy that has been proposed. A separate method has been developed specific for the analysis of ^{224}Ra and ^{226}Ra. The method involves collection of Ra on MnO_2, dissolution with hydrogen peroxide, co-precipitation with $PbSO_4$, dissolution with diethylene-triaminepentaacetic acid (DTPA) and co-precipitation with $BaSO_4$ followed by mounting for alpha spectrometry. ^{228}Ra has been determined via gamma spectrometric analysis of the daughter nuclide ^{228}Ac, assuming secular equilibrium has been established. The second method, which is still in development, will be utilized for analyzing the remaining analytes of interest. When implemented, the determination of ^{90}Sr and actinides will utilize a sequential strategy from the same aliquot of sample.

Results

Prior to analyses, brine samples were treated to ensure removal of residual hydrocarbon phase. All brine samples contained varying amounts of a red precipitate, thought to be an iron compound. To eliminate this precipitate, 2 mL of concentrated HCl per liter of brine was added. This addition was effective in removing most of the precipitate, however, in a few cases 2 mL of 30% H_2O_2 per liter of brine was added to ensure complete dissolution. All brine samples were filtered through 15 cm Whatman 41 ashless filter paper to remove residual hydrocarbon. Sufficient brine was filtered to fill a 3-L Marinelli beaker for gamma spectrometric analysis.

To date only the gamma-emitting radionuclides have been determined. Any additional analyses will be performed on aliquots of the same 3 L brine sample analyzed by gamma spectrometry. Gamma spectrometry results are presented in Table 22 for the 18 batteries sampled. Only ^{40}K and ^{228}Ac were detected in the samples. The activity for ^{228}Ac is indicative of the ^{228}Ra activity, as sufficient time had elapsed for the two radionuclides to be in secular equilibrium. No other gamma-emitting radionuclides were

detected in the samples. In all cases ^{137}Cs was below its minimal detectable concentration (~0.065 Bq/L) in the samples.

Table 22. Gamma Spectrometric Results for Brine Samples from 18 Batteries (Bq/L)

Battery	^{40}K	Uncertainty	MDC
Martha	5.90E+01	1.04E+00	1.08E+00
Graham	5.86E+01	1.02E+00	9.72E-01
Adeline	5.81E+01	1.02E+00	1.06E+00
Pauline	5.64E+01	9.91E-01	1.01E+00
Unocal	5.41E+01	9.71E-01	1.09E+00
Rosemary	5.77E+01	1.01E+00	9.97E-01
Flora	5.84E+01	1.01E+00	9.47E-01
Lost Tank	6.74E+01	1.15E+00	1.01E+00
Dolores	5.32E+01	9.37E-01	9.20E-01
Mary	5.10E+01	9.07E-01	9.17E-01
Jacque	5.83E+01	1.02E+00	9.72E-01
Lilly	5.71E+01	9.98E-01	9.75E-01
Kiwi	5.82E+01	1.13E+00	1.33E+00
Cleary	5.49E+01	9.63E-01	9.32E-01
Medano	5.11E+01	9.13E-01	9.86E-01
Wolf	5.34E+01	9.55E-01	1.05E+00
Bonneville	5.06E+01	8.98E-01	9.07E-01
Lucy	5.74E+01	1.01E+00	1.07E+00
Battery	**^{228}Ac**	**Uncertainty**	**MDC**
Martha	1.89E+01	3.16E-01	3.07E-01
Graham	1.88E+01	3.11E-01	2.55E-01
Adeline	1.64E+01	2.77E-01	2.79E-01
Pauline	2.25E+01	3.70E-01	2.96E-01
Unocal	2.29E+01	3.73E-01	2.13E-01
Rosemary	1.58E+01	2.67E-01	2.84E-01
Flora	1.09E+01	1.91E-01	2.34E-01
Lost Tank	1.75E+01	2.90E-01	2.01E-01
Dolores	1.68E+01	2.82E-01	2.77E-01
Mary	1.71E+01	2.87E-01	2.65E-01
Jacque	1.72E+01	2.86E-01	2.08E-01
Lilly	1.78E+01	2.98E-01	2.83E-01
Kiwi	1.80E+01	3.16E-01	2.86E-01
Cleary	1.91E+01	3.17E-01	2.93E-01
Medano	1.51E+01	2.52E-01	1.73E-01
Wolf	1.12E+01	1.98E-01	3.03E-01
Bonneville	1.91E+01	3.16E-01	2.74E-01
Lucy	1.92E+01	3.16E-01	1.99E-01
Battery	**$^{228}Ra^{*}$**	**Uncertainty**	**MDC**
Martha	1.89E+01	3.16E-01	3.07E-01
Graham	1.88E+01	3.11E-01	2.55E-01
Adeline	1.64E+01	2.77E-01	2.79E-01
Pauline	2.25E+01	3.70E-01	2.96E-01
Unocal	2.29E+01	3.73E-01	2.13E-01

Table continued on next page

Table 22. Gamma Spectrometric Results for Brine Samples from 18 Batteries (Bq/L) (cont.)

Battery	^{228}Ra[a]	Uncertainty	MDC
Rosemary	1.58E+01	2.67E-01	2.84E-01
Flora	1.09E+01	1.91E-01	2.34E-01
Lost Tank	1.75E+01	2.90E-01	2.01E-01
Dolores	1.68E+01	2.82E-01	2.77E-01
Mary	1.71E+01	2.87E-01	2.65E-01
Jacque	1.72E+01	2.86E-01	2.08E-01
Lilly	1.78E+01	2.98E-01	2.83E-01
Kiwi	1.80E+01	3.16E-01	2.86E-01
Cleary	1.91E+01	3.17E-01	2.93E-01
Medano	1.51E+01	2.52E-01	1.73E-01
Wolf	1.12E+01	1.98E-01	3.03E-01
Bonneville	1.91E+01	3.16E-01	2.74E-01
Lucy	1.92E+01	3.16E-01	1.99E-01

[a]Results for ^{228}Ra were determined from ^{228}Ac assuming secular equilibrium

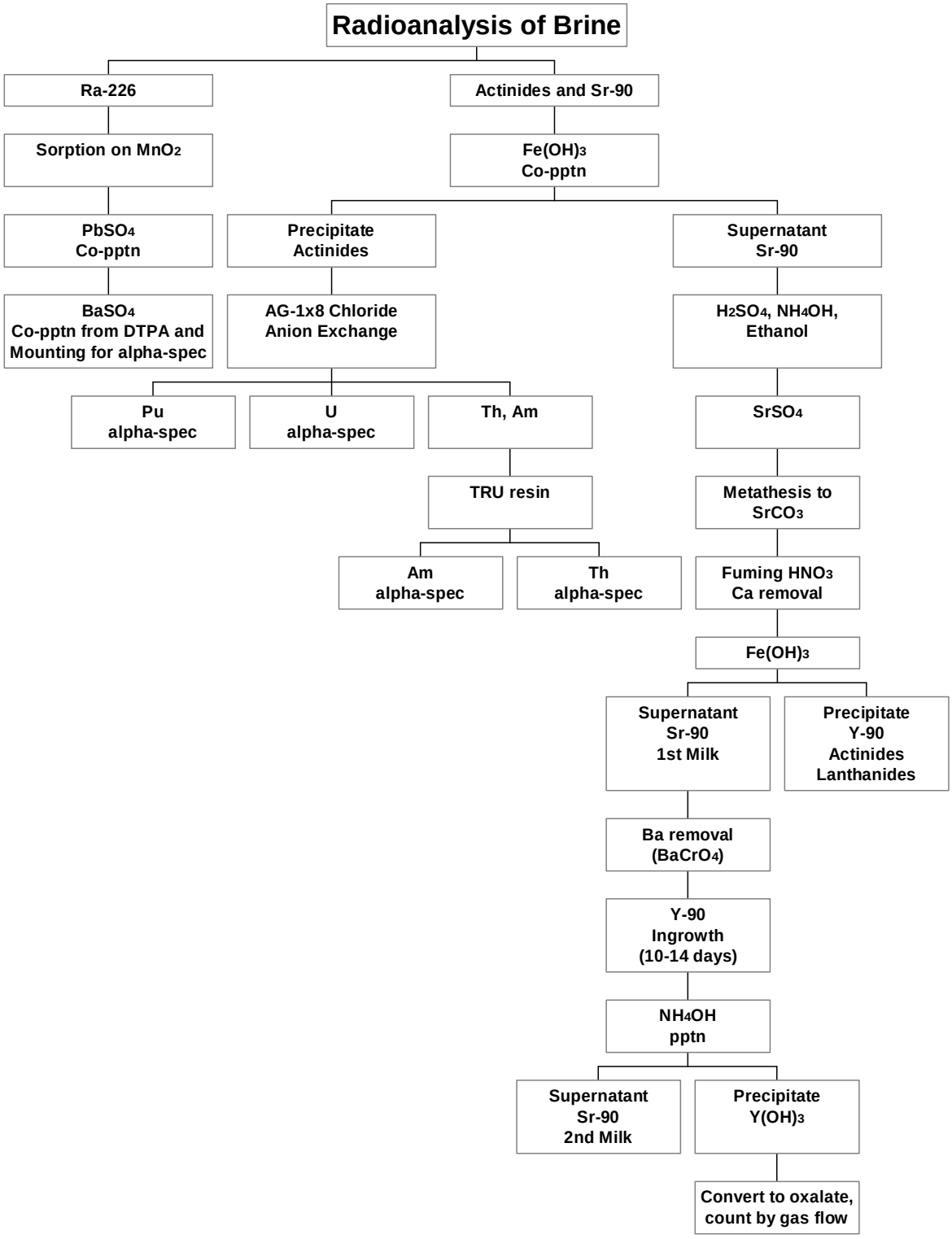

Figure 26. Analytical flowchart of radionuclide separation and analysis strategy for brine characterization

Radionuclides and Inorganics in Selected Drinking Water Sources

Introduction

The water wells in the immediate vicinity of the WIPP site provide water primarily for livestock, industrial usage by oil and gas production operations, and monitoring studies conducted by various groups. In April 2002, water samples were collected for CEMRC environmental monitoring studies from five sources in the region of the WIPP. The sources included the community water supplies of Carlsbad, Loving, Otis, and Hobbs; and the water supply for the WIPP site (Double Eagle). In past years a sixth source, a private well (Private Well #2), was sampled, however, since the 2001 sampling period this well has been dry, and thus it is likely no further samples will be obtained.

Aquifers in the region surrounding the WIPP include Dewey Lake, Culebra-Magenta, Ogalalla, Dockum, Pecos River alluvium and Capitan Reef. The main Carlsbad water supply is the Sheep Draw well field whose primary source is the Capitan Reef aquifer. The Hobbs and WIPP-Double Eagle water supplies are drawn from the Ogalalla aquifer, while the Loving/Malaga and Otis supply wells draw from deposits that are hydraulically linked to the flow of the Pecos River.

The 2001 and 2002 drinking water samples were collected after WIPP began receiving both radioactive waste (March 1999) and mixed waste (September 2000). Therefore, this summary represents continued monitoring phase data for radionuclides and non-radiological constituents in drinking water.

CEMRC began collecting drinking water samples in 1997, and summaries of methods, data and results from previous sampling were reported in the CEMRC 1997, 1998, 1999, 2000 and 2001 reports (available at http://www.cemrc.org).

Methods

All 2002 samples were collected according to EPA protocols for the collection, handling and preservation of drinking water as follows: (1) 4 L for radiological analyses, (2) 1 L for elemental analyses and (3) 1 L for anion tests. None of the samples were filtered before analysis, but a portion of the 4-L sample was transferred to a 3-L Marinelli beaker for gamma spectroscopy analyses. The 2002 samples were collected at the same locations as the 2001 samples.

CEMRC performed non-radiological analyses of drinking water samples using IC, ICP-MS and AAS.

Radiological analyses were carried out at CEMRC by first counting the samples in Marinelli beakers using a coaxial, high purity Ge detector system to determine gamma-emitting radionuclide activity concentrations. Radiochemistry was then applied to each sample for actinide separation and purification using multiple precipitation, co-precipitation and ion-exchange and/or extraction chromatography. Once the actinides were separated elementally, they were co-precipitated with NdF_3 and deposited onto filters, which were then counted on an alpha spectroscopy system.

Results and Discussion

Radiological

No naturally occurring or anthropogenic radionuclides (other than radon decay progeny) were measured above MDC in 2002 samples as determined by gamma spectroscopy. Three naturally occurring actinides (^{234}U, ^{235}U and ^{238}U) were detected via alpha spectroscopy in all of the samples from each location. However, the results for the quality control samples (trip and tracer blanks) analyzed with the 2002 drinking water samples indicated a potential uranium contamination problem in the laboratory. Although the source of the contamination was determined and eliminated, uranium results based on radiochemical methods for the 2002 samples were suspect. Additional samples were not readily available for re-analysis. A change will be instituted in the 2003 drinking water sampling protocol allowing for the collection of 8 L for radiological analyses (4 L for analysis and 4 L to be archived until all analyses are successfully completed). As an alternative to the alpha spectrometry results

for the 2002 samples, total uranium values obtained from ICP-MS analysis have been used to calculate isotopic concentrations and are presented for comparison with previous years. These values compare well with those measured in the past and also with the 2002 results (even with the potential uranium contamination problem) by alpha spectrometry. The relative percent difference between the calculated 2002 and measured 2001 values vary from 0.1 to 20% for the five sources sampled.

Calculated values for samples collected during 2002 were 27-135 $mBqL^{-1}$ for ^{234}U, 0.83-2.9 $mBqL^{-1}$ for ^{235}U, and 10-50 $mBqL^{-1}$ for ^{238}U. The highest levels of all three uranium isotopes for each year studied were in samples from Otis (Figs. 27-29). Across all years, ^{234}U concentrations were 3.2-3.4 times greater than ^{238}U in samples from Loving, and 2.3-2.8 times greater in samples from the other four drinking water sources. Enrichment of ^{234}U at these levels is common in drinking water produced from underground sources (Eisenbud and Gesell, 1997).

The ranges and ratios of all three uranium isotopes measured in CEMRC samples during 1998 - 2002 were similar to values from 1992 samples from Carlsbad, Double Eagle and Loving reported by EEG (Kenney, 1994).

The levels and ratios of uranium are typical of natural variations in ground water (Cothern, and Lappenbusch, 1983; Luo et al., 2000), and agree well with the few directly comparable values reported from studies in the region. Based on other relevant scientific information and results of analyses of other media reported herein, CEMRC concludes that the higher levels of some uranium isotopes in the 2002 drinking water samples from some sources represent a combination of natural variation and analytical artifact, and are not the result of releases from the WIPP.

Pu was not detected in any 2002 drinking water samples.

Non-Radiological Results

Measurements of inorganic analytes produced by CEMRC from the five drinking water sources showed little variation between years for each source. With a few exceptions the 1998-2002 measurements exhibit a high level of consistency that provides a useful characterization of each source (Tables 23-25). Hg was not detectable in samples from any of the sources this year. Measured As levels were highest in samples from Hobbs (4.9-7.45 μgL^{-1}) and Double Eagle (5.2-7.4 μgL^{-1}), and these measurements suggest that these drinking water sources would exceed the As standard $\leq$ 5 μgL^{-1} recently adopted by EPA (with full compliance required by 2006).

As in previous years, measured levels of chloride and sulfate exceeded reference levels (secondary maximum contaminant levels) in the 2002 samples for Otis.

These results are not appropriate for use in assessing of regulatory compliance, due to sample collection locations and other methodological details. However, it is noteworthy that the CEMRC results for Carlsbad and Double Eagle drinking water collected during 1998-2002 generally agreed well with measurements published by the City of Carlsbad Municipal Water System (*2002, 2001, 2000, 1999, 1998)* and with measurements published by the Otis Water User Co-Op (1999). As noted in previous reports, CEMRC values for nitrates are higher than those reported by Carlsbad and Loving, because the city-reported values are actually total N, rather than total nitrates. Based on these comparisons and the results of the analyses of the other media reported in this document, CEMRC concludes the higher levels of some inorganic analytes in the 2002 drinking water sources are not the result of releases from the WIPP.

Although a series of changes have been recently implemented for sampling and analysis of other environmental media under the WIPP-EM project, annual sampling of the same five sources as in prior years has been proposed for drinking water. This is in response to the 2002 public survey, which indicated that drinking water was the environmental medium of greatest concern.

Tables presenting drinking water data summarized herein are available on the CEMRC web site at http://www.cemrc.org.

Table 23. Range of Concentrations of Selected Inorganic Analytes in Drinking Water Samples Collected from Carlsbad and Double Eagle during 1998-2002

Analyte	Unit	Carlsbad					Double Eagle				
		1998-2000			[d]Conc. 2001	Conc. 2002	1998-2000				
		[a]N	[b]Min	[c]Max			N	Min	Max	Conc. 2001	Conc. 2002
Ag	µg/L	1	1.75E-02	1.75E-02	[e]NA	NA	0	NA	NA	NA	NA
Al	µg/L	2	2.34E+00	3.17E+01	3.68E+00	NA	3	6.76E-01	7.22E+01	6.08E+00	4.30E+00
As	µg/L	3	5.67E-01	6.82E-01	NA	3.45E-01	3	5.21E+00	7.42E+00	5.60E+00	6.31E+00
Ba	µg/L	3	6.83E+01	6.96E+01	6.64E+01	6.98E+01	2	7.97E+01	8.96E+01	1.25E+02	9.37E+01
Be	µg/L	0	NA	NA	NA	NA	1	3.63E-02	3.63E-02	NA	NA
Ca	µg/L	3	7.26E+04	7.98E+04	8.06E+04	7.00E+04	3	5.08E+03	5.83E+04	5.11E+04	5.61E+04
Cd	µg/L	0	NA	NA	NA	NA	0	NA	NA	NA	NA
Ce	µg/L	0	NA	NA	NA	NA	0	NA	NA	3.63E-03	NA
Co	µg/L	2	1.83E-01	3.41E-01	8.80E-02	2.15E-01	2	9.31E-02	1.37E-01	NA	1.12E+00
Cr	µg/L	2	2.68E+00	4.01E+00	3.81E+00	6.96E+00	2	2.94E+00	3.20E+00	1.29E+00	4.98E+00
Cu	µg/L	2	1.81E+00	4.29E+00	6.90E+00	1.30E+00	2	1.19E+00	4.84E+00	2.54E+00	8.09E-01
Dy	µg/L	0	NA	NA	NA	NA	0	NA	NA	NA	NA
Eu	µg/L	2	1.80E-02	2.42E-02	1.35E-02	2.12E-02	2	1.88E-02	2.58E-02	2.70E-02	2.86E-02
Fe	µg/L	1	2.07E+00	2.07E+00	NA	2.24E+01	2	1.98E+00	7.93E+01	1.48E+02	NA
Ga	µg/L	0	[f]ND	ND	ND	3.25E+00	0	ND	ND	ND	4.46E+00
Gd	µg/L	0	NA	NA	NA	NA	0	NA	NA	NA	NA
Hg	µg/L	0	NA	NA	NA	NA	0	NA	NA	NA	NA
K	µg/L	3	1.27E+03	2.91E+03	3.56E+03	1.04E+03	3	2.66E+03	2.94E+04	2.62E+03	2.51E+03
La	µg/L	2	1.35E-02	4.42E-02	NA	1.85E-02	2	1.59E-02	6.26E-02	1.19E-02	2.47E-02
Li	µg/L	2	7.25E+00	7.87E+00	7.41E+00	6.09E+00	3	1.81E+01	1.90E+01	1.37E+01	1.51E+01
Mg	µg/L	2	3.14E+04	3.40E+04	3.43E+04	3.14E+04	2	1.09E+03	1.07E+04	9.21E+03	9.54E+03
Mn	µg/L	3	5.50E-02	3.37E-01	9.40E-01	1.16E-01	2	2.30E-01	3.52E-01	8.73E-01	2.93E-01
Mo	µg/L	3	7.03E-01	1.20E+00	8.93E-01	1.14E+00	2	1.42E+00	1.66E+00	1.48E+00	1.66E+00
Na	µg/L	3	1.97E+04	9.94E+04	4.55E+04	1.29E+04	3	3.84E+03	3.25E+04	2.35E+04	2.73E+04
Nd	µg/L	0	NA	NA	NA	NA	0	NA	NA	NA	NA
Ni	µg/L	2	2.13E+00	2.84E+00	1.46E+00	2.01E+00	2	1.44E+00	1.72E+00	1.66E+00	1.28E+00
Pb	µg/L	2	3.76E-01	1.44E+00	6.32E-01	1.68E-01	2	3.16E-01	1.38E+00	6.92E-01	2.56E-01
Pr	µg/L	0	NA	NA	NA	NA	0	NA	NA	NA	NA
Rh	µg/L	0	ND	ND	ND	NA	0	ND	ND	ND	1.56E-02
Sb	µg/L	1	1.99E-01	1.99E-01	NA	3.60E-02	3	2.41E-02	1.39E-01	NA	3.71E-02
Sc	µg/L	1	1.82E+00	1.82E+00	1.57E+00	3.03E+00	1	5.27E+00	5.27E+00	4.61E+00	6.59E+00
Se	µg/L	0	NA	NA	NA	1.22E+00	0	NA	NA	3.41E+00	3.53E+00
Si	µg/L	0	ND	ND	ND	6.87E+03	0	ND	ND	ND	1.81E+04
Sm	µg/L	2	2.34E-02	2.57E-02	3.39E-02	3.61E-02	2	2.83E-02	3.70E-02	3.31E-02	4.26E-02
Sn	µg/L	0	NA	NA	5.97E-02	NA	1	3.36E-01	3.36E-01	NA	NA
Sr	µg/L	3	3.53E+02	4.59E+02	3.59E+02	3.23E+02	3	5.06E+01	5.28E+02	5.53E+02	5.43E+02
Th	µg/L	1	1.76E-02	1.76E-02	NA	NA	1	2.07E-03	2.07E-03	NA	NA
Ti	µg/L	0	NA	NA	1.60E+00	NA	0	NA	NA	2.87E+00	NA
Tl	µg/L	2	1.11E-01	1.20E-01	1.54E-01	1.24E-01	0	NA	NA	NA	NA

Table continued on next page

Table 23. Range of Concentrations of Selected Inorganic Analytes in Drinking Water Samples Collected from Carlsbad and Double Eagle during 1998-2002 (cont.)

Analyte	Unit	Carlsbad					Double Eagle				
		1998-2000			[d]Conc. 2001	Conc. 2002	1998-2000				
		[a]N	[b]Min	[c]Max			N	Min	Max	Conc. 2001	Conc. 2002
U	µg/L	2	8.21E-01	8.42E-01	8.43E-01	8.49E-01	2	1.75E+00	1.77E+00	1.48E+00	1.34E+00
V	µg/L	3	3.82E+00	4.69E+00	4.59E+00	5.80E+00	2	2.65E+01	2.70E+01	2.80E+01	3.26E+01
Zn	µg/L	3	4.52E+00	1.52E+01	7.11E+00	4.39E+00	2	1.46E+00	5.19E+00	1.25E+01	1.80E+00
Chloride	µg/L	3	1.53E+04	1.88E+05	6.65E+04	1.32E+04	3	2.58E+04	3.69E+04	2.43E+04	2.40E+04
Fluoride	µg/L	3	2.18E+02	7.81E+02	5.36E+02	5.72E+02	3	5.01E+02	9.71E+02	7.86E+02	8.37E+02
Nitrate	µg/L	3	3.52E+03	5.91E+03	4.06E+03	3.83E+03	3	1.07E+04	1.36E+04	1.20E+04	1.17E+04
Phosphate	µg/L	0	NA	NA	NA	NA	0	NA	NA	NA	NA
Sulfate	µg/L	3	8.07E+04	1.17E+05	9.96E+04	7.71E+04	3	4.12E+04	5.69E+04	4.09E+04	4.14E+04

[a]N = number of samples > MDL (1998-2000)
[b]Min = minimum measured concentration in annual samples from 1998-2000
[c]Max = maximum measured concentration in annual samples from 1998-2000
[d]Conc. = concentration
[e]NA = all samples below MDL for analyte
[f]ND = Not analyzed in samples before 2002

Table 24. Range of Concentrations of Selected Inorganic Analytes in Drinking Water Samples Collected from Hobbs and Loving during 1998-2002

Analyte	Unit	Hobbs					Loving				
		1998-2000			[d]Conc. 2001	Conc. 2002	1998-2000				
		[a]N	[b]Min	[c]Max			N	Min	Max	Conc. 2001	Conc. 2002
Ag	µg/L	1	3.86E-03	3.86E-03	[e]NA	1.04E-01	2	2.55E-03	3.28E-03	NA	1.30E-01
Al	µg/L	3	3.03E+00	1.14E+02	8.62E+00	8.01E+00	2	1.30E+00	1.56E+00	3.76E+00	NA
As	µg/L	3	5.71E+00	7.37E+00	4.91E+00	6.85E+00	3	1.48E+00	1.85E+00	1.67E+00	1.20E+00
Ba	µg/L	3	5.65E+01	5.99E+01	5.98E+01	5.83E+01	3	2.90E+01	3.03E+01	3.01E+01	2.98E+01
Be	µg/L	1	5.39E-02	5.39E-02	NA	NA	1	9.35E-02	9.35E-02	NA	NA
Ca	µg/L	3	7.99E+03	9.06E+04	7.79E+04	8.66E+04	3	9.04E+03	1.04E+05	9.02E+04	1.00E+05
Cd	µg/L	1	4.34E-03	4.34E-03	NA	NA	0	NA	NA	NA	NA
Ce	µg/L	3	5.87E-03	2.23E-02	5.10E-03	NA	1	9.74E-04	9.74E-04	NA	NA
Co	µg/L	3	2.01E-01	3.61E-01	9.78E-02	2.89E-01	3	1.48E-01	2.48E-01	1.02E-01	4.04E-01
Cr	µg/L	3	2.28E+00	3.10E+00	1.18E+00	5.20E+00	3	2.51E+00	4.28E+00	1.91E+00	7.44E+00
Cu	µg/L	3	1.87E+00	2.70E+00	1.06E+00	1.69E+00	3	2.35E+00	5.59E+00	2.11E+00	2.36E+00
Dy	µg/L	0	NA	NA	4.18E-03	NA	0	NA	NA	NA	NA
Eu	µg/L	3	1.37E-02	1.97E-02	1.31E-02	1.78E-02	3	7.00E-03	1.01E-02	7.92E-03	8.91E-03
Fe	µg/L	2	2.10E+00	6.70E+01	1.74E+02	NA	0	NA	NA	2.24E+02	3.13E+01
Ga	µg/L	0	[f]ND	ND	ND	2.56E+00	0	ND	ND	ND	1.26E+00
Gd	µg/L	0	NA	NA	NA	NA	1	2.15E-03	2.15E-03	NA	NA
Hg	µg/L	2	1.06E-02	1.42E-02	NA	NA	0	NA	NA	NA	NA
K	µg/L	3	2.44E+03	2.52E+04	2.59E+03	2.50E+03	2	1.95E+03	2.45E+03	2.13E+03	2.04E+03
La	µg/L	3	1.45E-02	5.01E-02	NA	1.67E-02	3	6.66E-03	2.22E-02	NA	7.31E-03
Li	µg/L	3	2.92E+01	3.18E+01	2.65E+01	2.97E+01	3	1.85E+01	1.96E+01	1.72E+01	1.87E+01
Mg	µg/L	2	2.11E+03	2.00E+04	1.92E+04	2.00E+04	2	4.04E+03	3.83E+04	3.88E+04	4.02E+04
Mn	µg/L	3	3.79E-01	6.69E-01	6.73E-01	5.09E-01	3	1.43E-02	8.59E-02	6.24E-02	5.83E-02
Mo	µg/L	3	2.46E+00	2.72E+00	3.07E+00	2.94E+00	3	1.28E+00	1.57E+00	1.58E+00	1.54E+00
Na	µg/L	3	4.97E+03	4.75E+04	4.24E+04	4.83E+04	3	2.33E+03	2.11E+04	1.96E+04	2.29E+04
Nd	µg/L	3	3.01E-03	1.28E-02	NA	NA	1	3.37E-03	3.37E-03	NA	NA
Ni	µg/L	3	2.22E+00	2.64E+00	1.67E+00	1.82E+00	3	2.71E+00	3.38E+00	2.03E+00	2.83E+00
Pb	µg/L	3	9.82E-02	1.70E-01	9.44E-02	1.44E-01	3	1.03E+00	1.67E+00	1.01E+00	9.40E-01
Pr	µg/L	1	1.57E-03	1.57E-03	NA	NA	0	NA	NA	NA	NA
Rh	µg/L	0	ND	ND	ND	2.52E-02	0	ND	ND	ND	3.07E-02
Sb	µg/L	2	3.88E-02	7.02E-02	6.15E-02	6.04E-02	2	6.78E-02	1.84E-01	NA	3.51E-02
Sc	µg/L	1	8.58E+00	8.58E+00	7.17E+00	1.01E+01	1	3.22E+00	3.22E+00	3.40E+00	4.72E+00
Se	µg/L	0	NA	NA	3.50E+00	6.23E+00	0	NA	NA	NA	NA
Si	µg/L	0	ND	ND	ND	2.86E+04	0	ND	ND	ND	1.09E+04
Sm	µg/L	3	2.20E-02	3.02E-02	1.93E-02	3.03E-02	3	8.43E-03	1.30E-02	NA	NA
Sn	µg/L	0	NA	NA	NA	NA	1	4.45E-01	4.45E-01	NA	NA
Sr	µg/L	3	7.89E+01	9.23E+02	8.86E+02	1.04E+03	3	7.60E+01	8.32E+02	8.55E+02	9.37E+02
Th	µg/L	2	2.29E-03	4.56E-03	NA	NA	2	5.69E-03	7.38E-03	NA	NA
Ti	µg/L	0	NA	NA	4.64E+00	3.14E+00	0	NA	NA	2.68E+00	5.91E+00
Tl	µg/L	0	NA	NA	2.24E-02	NA	0	NA	NA	4.32E-02	NA
U	µg/L	3	2.90E+00	3.39E+00	3.23E+00	2.99E+00	3	1.98E+00	2.11E+00	2.26E+00	2.10E+00
V	µg/L	3	3.40E+01	3.56E+01	3.63E+01	3.71E+01	3	1.22E+01	1.27E+01	1.20E+01	1.44E+01
Zn	µg/L	3	8.44E-01	4.37E+00	2.06E+00	1.58E+00	3	4.79E+00	9.03E+00	9.01E+00	5.74E+00

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Table 24. Range of Concentrations of Selected Inorganic Analytes in Drinking Water Samples Collected from Hobbs and Loving during 1998-2002 (cont.)

Analyte	Unit	Hobbs					Loving				
		1998-2000			[d]Conc. 2001	Conc. 2002	1998-2000				
		[a]N	[b]Min	[c]Max			N	Min	Max	Conc. 2001	Conc. 2002
Chloride	µg/L	3	6.32E+04	9.36E+04	7.40E+04	7.46E+04	3	1.59E+04	2.94E+04	2.50E+04	2.74E+04
Fluoride	µg/L	3	6.19E+02	1.33E+03	1.15E+03	1.16E+03	3	2.45E+02	6.45E+02	6.10E+02	NA
Nitrate	µg/L	3	1.70E+04	2.01E+04	1.58E+04	1.56E+04	3	1.72E+04	2.32E+04	1.66E+04	1.59E+04
Phosphate	µg/L	0	NA	NA	NA	NA	0	NA	NA	NA	NA
Sulfate	µg/L	3	1.04E+05	1.42E+05	9.60E+04	1.06E+05	3	1.45E+05	2.05E+05	1.56E+05	1.54E+05

[a]N = number of samples > MDL (1998-2000)
[b]Min = minimum measured concentration in annual samples from 1998-2000
[c]Max = maximum measured concentration in annual samples from 1998-2000
[d]Conc. = concentration
[e]NA = all samples below MDL for analyte
[f]ND = Not analyzed in samples before 2002

Table 25. Range of Concentrations of Selected Inorganic Analytes in Drinking Water Samples Collected from Otis during 1998-2002

Analyte	Unit	Otis				
		1998-2000			Concentration 2001	Concentration 2002
		[a]N	[b]Min	[c]Max		
Ag	µg/L	1	2.63E-02	2.63E-02	NA	NA
Al	µg/L	1	1.42E+00	1.42E+00	NA	5.74E+00
As	µg/L	3	1.30E+00	1.56E+00	NA	6.53E-01
Ba	µg/L	3	1.39E+01	1.70E+01	1.38E+01	1.75E+01
Be	µg/L	0	NA	NA	NA	NA
Ca	µg/L	3	2.14E+05	3.17E+05	3.83E+05	2.81E+05
Cd	µg/L	0	NA	NA	NA	NA
Ce	µg/L	0	NA	NA	NA	NA
Co	µg/L	2	3.92E-01	6.82E-01	3.21E-01	9.51E-01
Cr	µg/L	3	1.11E+00	4.08E+00	1.41E+00	6.67E+00
Cu	µg/L	2	5.51E+00	6.02E+00	3.42E+00	4.50E+00
Dy	µg/L	0	NA	NA	3.39E-03	NA
Eu	µg/L	1	3.42E-03	3.42E-03	4.73E-03	9.48E-03
Fe	µg/L	2	2.87E+00	2.99E+01	8.53E+02	1.10E+02
Ga	µg/L	0	ND	ND	ND	6.54E-01
Gd	µg/L	0	NA	NA	NA	NA
Hg	µg/L	0	NA	NA	NA	NA
K	µg/L	3	2.74E+03	3.93E+03	4.01E+03	3.32E+03
La	µg/L	1	3.36E-03	3.36E-03	NA	6.30E-03
Li	µg/L	2	4.70E+01	4.85E+01	4.51E+01	4.11E+01
Mg	µg/L	2	7.95E+04	8.83E+04	1.08E+05	9.58E+04
Mn	µg/L	2	2.00E-01	2.53E-01	1.78E-01	2.32E+00
Mo	µg/L	2	2.25E+00	2.75E+00	2.49E+00	2.53E+00
Na	µg/L	3	7.83E+04	1.14E+05	1.62E+05	1.30E+05
Nd	µg/L	1	4.80E-03	4.80E-03	3.97E-02	1.36E-02
Ni	µg/L	2	7.17E+00	1.06E+01	6.23E+00	8.94E+00
Pb	µg/L	2	1.08E-01	1.91E-01	1.19E-01	5.04E-01
Pr	µg/L	0	NA	NA	NA	NA
Rh	µg/L	0	ND	ND	ND	1.29E-01
Sb	µg/L	2	5.03E-02	4.10E-01	NA	3.69E-02
Sc	µg/L	1	3.53E+00	3.53E+00	4.68E+00	4.28E+00
Se	µg/L	0	NA	NA	NA	NA
Si	µg/L	0	ND	ND	ND	1.04E+04
Sm	µg/L	1	3.56E-03	3.56E-03	NA	NA
Sn	µg/L	0	NA	NA	NA	NA
Sr	µg/L	3	2.38E+03	2.86E+03	3.61E+03	3.33E+03

Table continued on next page

Table 25. Range of Concentrations of Selected Inorganic Analytes in Drinking Water Samples Collected from Otis during 1998-2002 (cont.)

Analyte	Unit	Otis				
		1998-2000			Concentration 2001	Concentration 2002
		[a]N	[b]Min	[c]Max		
Th	µg/L	2	1.19E-03	2.67E-02	NA	NA
Ti	µg/L	0	NA	NA	5.68E+00	3.41E+01
Tl	µg/L	0	NA	NA	NA	NA
U	µg/L	2	3.95E+00	4.20E+00	5.34E+00	4.08E+00
V	µg/L	3	1.10E+01	1.17E+01	1.14E+01	1.29E+01
Zn	µg/L	3	4.39E+00	1.16E+01	1.64E+01	4.93E+00
Chloride	µg/L	3	1.36E+05	3.86E+05	4.21E+05	3.31E+05
Fluoride	µg/L	3	1.29E+02	5.52E+02	7.53E+02	NA
Nitrate	µg/L	3	9.59E+03	2.20E+04	1.90E+04	1.63E+04
Phosphate	µg/L	0	NA	NA	NA	NA
Sulfate	µg/L	3	3.27E+05	7.55E+05	7.56E+05	6.61E+05

[a]N = number of samples > MDL (1998-2000)
[b]Min = minimum measured concentration in annual samples from 1998-2000
[c]Max = maximum measured concentration in annual samples from 1998-2000
[d]NA = all samples below MDL for analyte
[e]ND = Not analyzed in samples before 2002

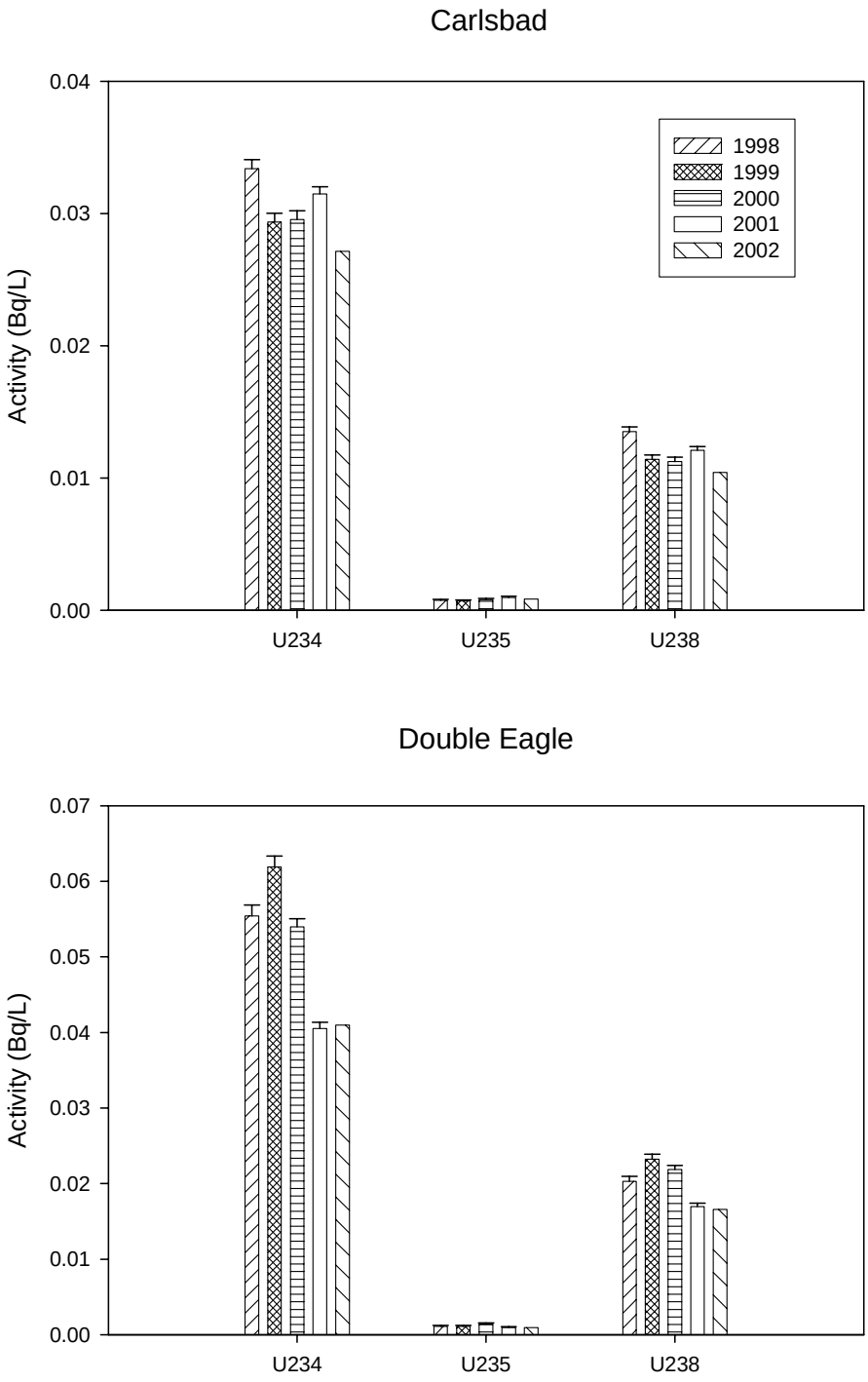

Figure 27. Activity Concentrations of Uranium Isotopes in Drinking Water Samples Collected at Carlsbad and Double Eagle during 1998-2002

Error bars represent the total radioanalytical uncertainty at 1 sigma. Note: Isotopic uranium results for ***2***002 samples were calculated from total uranium values obtained by ICP-MS analysis. Error bars have not been provided.

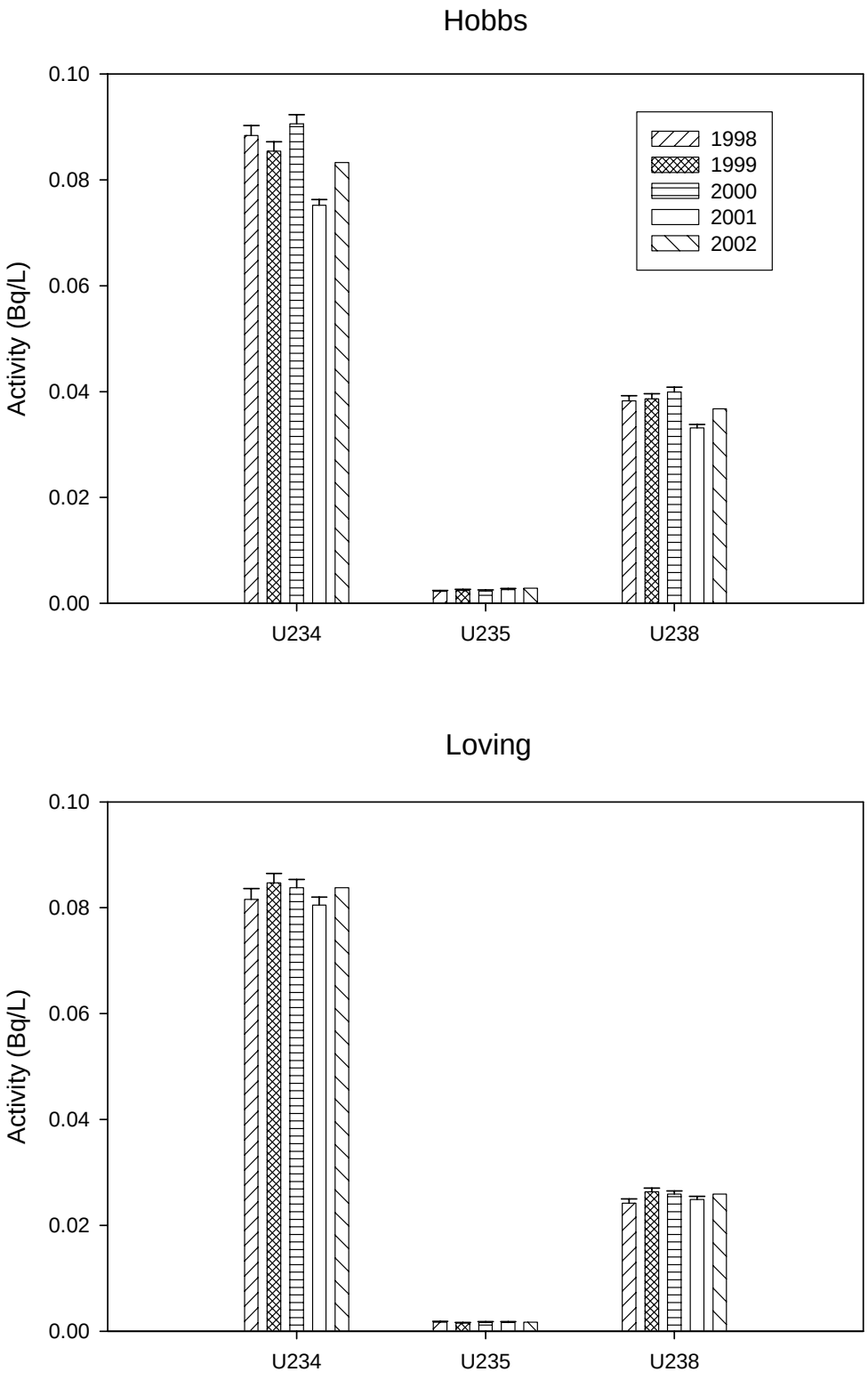

Figure 28. Activity Concentrations of Uranium Isotopes in Drinking Water Samples Collected at Hobbs and Loving during 1998-2002

Error bars represent the total radioanalytical uncertainty at 1 sigma. Note: Isotopic uranium results for 2002 samples were calculated from total uranium values obtained by ICP-MS analysis. Error bars have not been provided.

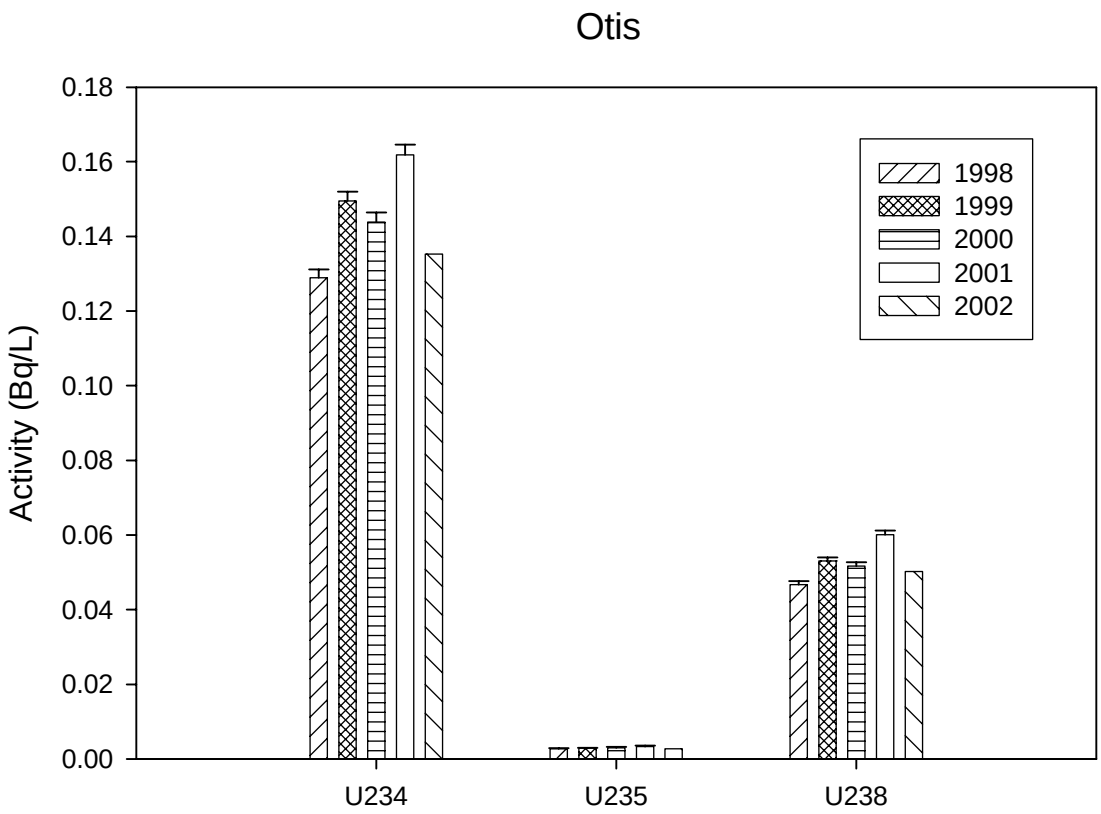

Figure 29. Activity Concentrations of Uranium Isotopes in Drinking Water Samples Collected at Otis during 1998-2002

Error bars represent the total radioanalytical uncertainty at 1 sigma. Note: Isotopic uranium results for 2002 samples were calculated from total uranium values obtained by ICP-MS analysis. Error bars have not been provided.

Actinide Chemistry

The Actinide Chemistry Team of LANL was established in Carlsbad under the Actinide Chemistry Program to provide the DOE with scientific and experimental support in the areas of actinide chemistry and repository science as they relate to the WIPP. The actinide team is located at CEMRC and works collaboratively with CEMRC staff. Experimental studies can enhance the credibility of the Performance Assessment model for the long-term disposal of radioactive waste. Confirmatory testing of various assumptions about the behavior of actinides under specific repository conditions provides better stakeholder acceptance, allows anticipation of issues before they arise in the public, and supports changes in the operations envelope of WIPP in the future. For the last two years, work has concentrated on the effects of radiolysis, backfill, and Fe/Al and other factors, on brine and actinide chemistry. Some of this work is summarized in the following sections. This program will respond to new research areas defined by DOE as they develop.

Goals of the LANL/CEMRC Actinide Chemistry Program in the next year include:

- Develop the laboratory resources and infrastructure in Carlsbad to safely conduct research with actinide elements.
- Establish a long-term experimental program to (1) more completely define the source term of actinides under the conditions expected in the WIPP, and (2) address operational needs as they arise such as head space gas generation and waste characterization.
- Conduct basic research in the field of actinide subsurface chemistry.
- Work with CEMRC/NMSU to develop educational and training programs in actinide and nuclear chemistry that target graduate and undergraduate students to help fulfill the future personnel needs of the DOE.

Pu can exist in more oxidation states in the environment than any other element. Oxidation state, which can be thought of as the excess charge on the central atom, for Pu can be +3, +4, +5, or +6 (designated by III, IV, V or VI). The fact that Pu species with various oxidation states (greater than 2) can co-exist in solution at the same time makes Pu chemistry very complicated and difficult to investigate. The development of laboratory resources and infrastructure is a key first step to begin investigations of such difficult systems, and to insure that facilities exist to properly support planned research with radioactive materials. Operationally, the infrastructure includes counting equipment, a defined and managed material balance area for inventory management and control, and laboratory work rules for conducting work with actinide elements.

Lastly, it remains an important goal in the Actinide Chemistry Program to coordinate efforts between LANL, CEMRC, WTS, NMSU and other educational institutions to develop needed graduate and undergraduate programs in actinide chemistry that will support DOE missions in the future. This is currently in the planning stages and will take a number of years to fully develop. Planned areas of development include training to work with radioactive materials, counting methods and analytical techniques; the development of courses in actinide chemistry, radiochemistry and the environmental chemistry of actinide systems; and the active pursuit of student and graduate level programs/participation at the CEMRC.

Specifically, work over the next few years will include continuation and expansion of current experiments including:

1. Study of the influence of major products of brine radiolysis on the plutonium (neptunium and uranium) oxidation states.
2. Development of a rapid and relatively easy method for determination of plutonium oxidation states in WIPP brines.
3. Investigations of the effect of metals (Fe/Al) on the oxidation states of plutonium (neptunium and uranium).
4. Determination of generation rates and stabilities of the major products of brine radiolysis (hydrogen peroxide,

hypochlorite, hydrogen) as a function of absorbed dose of alpha radiation.
5. Study of the effects of MgO Backfill on plutonium and brine chemistry.
6. Investigation of the pathways of hydrogen generation in low-level actinide wastes.

No results obtained, thus far, indicate any problems with the WIPP repository performance.

In a nuclear waste repository such as WIPP, the radiation from the wastes will produce hydrogen peroxide (H_2O_2) and hypochlorite (ClO^-) as radiolytic by-products in brine solution. Hydrogen peroxide, which is the most abundant radiolytic by-product in brine at high pH, acts like an efficient reductant (actually an electron donor), and can reduce Pu(VI), existing in solution as PuO_2^{2+} in brines, to the less mobile Pu(IV). The second by-product - hypochlorite - is an oxidant and can oxidize Pu(IV) up to Pu(VI). Competition between oxidizing and reducing conditions will have an influence on speciation and mobility of the actinides. Understanding this competition is a major focus of actinide research.

Although the Actinide Chemistry Program has only just relocated to CEMRC this year, some preliminary work was begun in 2002 at CEMRC on the effects of MgO Backfill. Sandia, LANL and CEMRC scientists in Carlsbad are currently engaged in an effort to better understand the role of backfill materials in geologic repositories of radioactive waste. A significant criterion in evaluating disposal strategies for radioactive waste is the assessment of the isolation capacity for the most radiotoxic radionuclides, the actinides. For some radioactive waste destined for disposal in geological salt deposits, such as WIPP, MgO is being used as a reactive backfill material. This material is ideal for buffering the pH of the repository to between 8.5 and 10, and for sorbing carbon dioxide and water, conditions that are favorable for stabilizing Pu in the repository. MgO can also serve to stabilize Pu through direct sorption processes.

Equilibration experiments with Pu, MgO Backfill, and brines were conducted at CEMRC and at Los Alamos. The brines included the WIPP brines ERDA-6 (brine E), GWBrine (brine G), 5 mole/L NaCl and 3.5 mole/L $MgCl_2 \cdot 6H_2O$. MgO-to-water ratios were varied to represent various scenarios of infiltration into the repository, e.g., 1:10, 1:0.25 and 1:0.15, to represent traditional batch tests, a fully indurated repository, and a partially indurated repository, respectively. At Los Alamos Pu was sorbed onto the MgO and subsequently desorbed using Pu-free brine to represent future indurations of brine into the repository. Release of Pu from Pu-loaded MgO agglomerates was determined in the presence or absence of hypochlorite (OCl^-), and under agitated and non-agitated conditions. After the Pu(VI)-brines were equilibrated with MgO Backfill for 68 days, the solution pH and alkalinity changed dramatically, while approximately 99% to 100% of the introduced Pu(VI) was removed from the brines. Under agitated conditions, the release rate of Pu from the Pu-loaded MgO agglomerates follows the order of MgO-Brine G > MgO-Brine E > MgO-NaCl > MgO-$MgCl_2 \cdot 6H_2O$. Under non-agitated conditions, a small amount of Pu was released from the Pu-loaded MgO-Brine G agglomerates after 110 days, but there was no Pu released from the Pu-loaded MgO-Brine E agglomerates (Fig. 30). Brine E is the brine of most significance to possible Pu releases and the performance of WIPP.

X-Ray Absorption Spectroscopy (XAS) results for the Pu-loaded MgO-agglomerates indicate that the Pu was incorporated by precipitation, and the oxidation state of the Pu was primarily Pu(IV) (Fig. 31). Unfortunately, the low levels of Pu used in these experiments made for low signal to noise ratios. However, reduction of Pu(VI) to Pu(IV) requires a reductant not just a pH change. This reductant comes from impurities in the MgO Backfill, another benefit of this particular material. Separate experiments in these brines with pH adjustment without MgO indicated that pH change alone could not account for the removal of Pu from Brine G solutions, but pH change alone could account for the removal of Pu from Brine E solutions. Therefore, in NaCl brines, like Brine E (ERDA-6), the buffering capability of the MgO Backfill alone may be adequate to completely retard Pu migration, while in MgCl

brines, like Brine G (GWBrine), the backfill is performing a sorptive function in addition to simple pH buffering that enhances its ability to retard Pu migration and enhances the performance of WIPP. Furthermore, XRD results performed at CEMRC on infusion of MgCl brines, like Brine G (GWBrine), showed complex phase formation under the low water:solid ratios expected in WIPP (Fig. 32) under repository types of infiltration. Phases like tachyhydrate ($CaMg_2Cl_6 \cdot 12\ H_2O$) have the potential to incorporate Pu into its crystal structure and form a stable host phase for actinides, e.g., $Ca_{1-x}(Pu,U,Th)_xMg_2Cl_6 \cdot 12H_2O$.

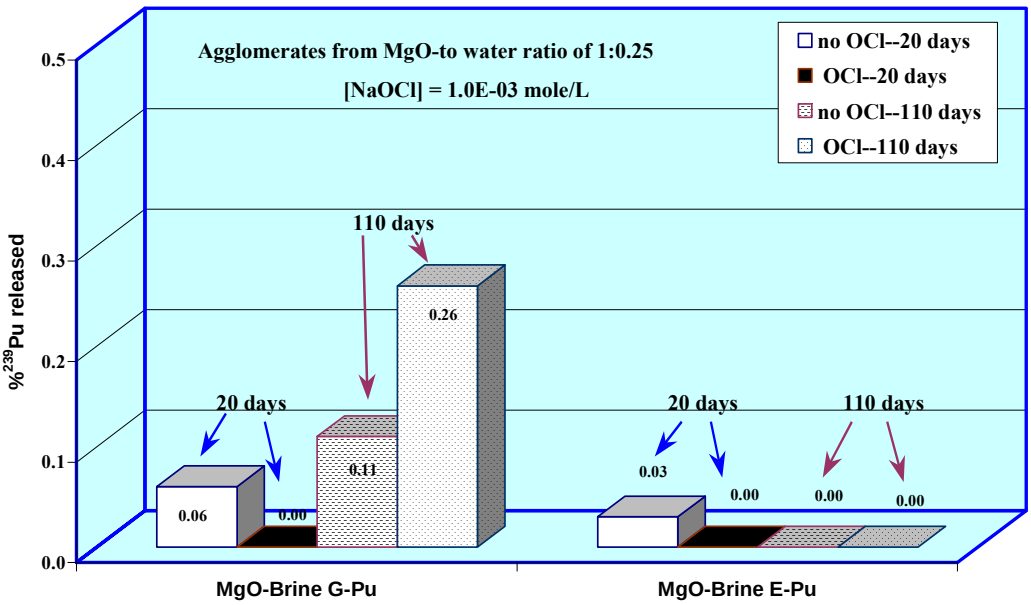

Figure 30. Desorption of ^{239}Pu from Pu-loaded MgO-Brine E and Brine G agglomerates under non-agitated conditions.

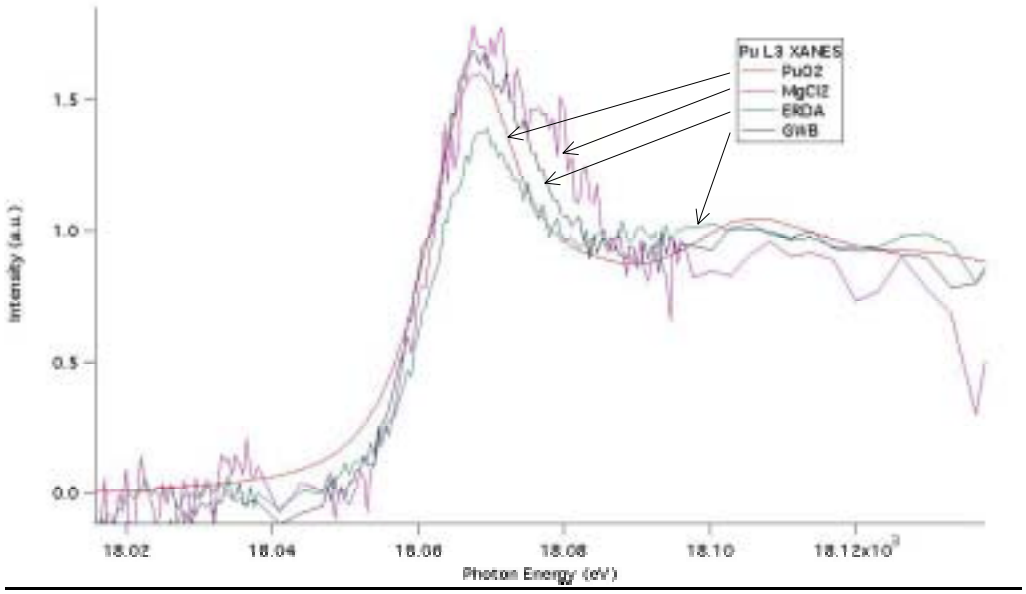

Figure 31. X-ray Absorption Spectra (XAS) of Pu in MgO--brine reaction products six months after the addition of the ^{239}Pu(VI), indicating all Pu as Pu(IV).

Note: XAS performed at Argonne National Laboratory.

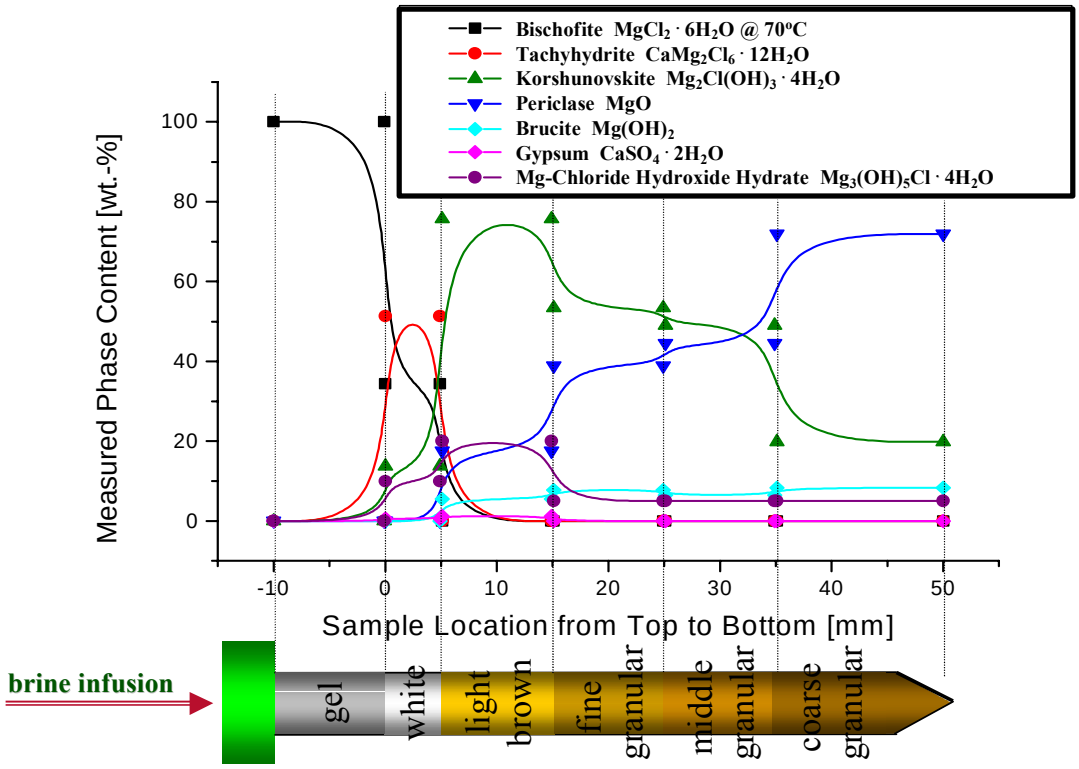

Figure 32. Phase Constitution of WIPP MgO Backfill after infiltration with 3.7 mole/L $MgCl_2$ brine and reaction for several weeks.

The contents of the individual identified phases were quantified by Rietveld refinement of X-ray powder data performed at CEMRC. The specific sample position from sample top (0 mm) and sample bottom (50 mm) are marked. The absolute content of Magnesium Chloride Hydroxide Hydrate ($Mg_3(OH)_5Cl \cdot 4H_2O$) is only roughly estimated since crystal structure data for this phase are not available.

Occurrence of Radionuclides in Residents of the Carlsbad, New Mexico Area and Public Awareness of the Center's WIPP Environmental Monitoring Program

Introduction

Citizen volunteers from the Carlsbad, New Mexico area were monitored for internally deposited radionuclides through a project entitled "Lie Down and Be Counted" (LDBC). This project is provided as an outreach service to the public and to support education about naturally occurring and man-made radioactivity present in people who live in the vicinity of the WIPP. The data collected prior to the opening of the WIPP facility (26 March 1999) serve as a baseline for comparisons with periodic follow-up measurements that are slated to continue throughout the 35-year operational phase of the WIPP. It is important to note that these data represent an interim summary (through 1 October 2002) of an ongoing study.

Participating in the LDBC consists of a lung and whole body count every two years. Volunteers are recruited through presentations to local community groups and businesses. The entire measurement process takes approximately one hour. A detailed description of the measurement protocol, analysis and instrument detection limits is provided in the CEMRC 1998 Report. In addition, the status of the project and results are updated quarterly on the CEMRC website (http://www.cemrc.org).

A survey was also conducted between February 25, 2002 and March 5, 2002 to assess the public awareness of the Center's WIPP environmental monitoring program, perceived risk posed by the WIPP site and what environmental media the public feels is important to monitor. The people surveyed included 158 people who have been counted at least twice under the Lie Down and Be Counted program.

Bioassay Results

As of 1 October 2002, 634 individuals had participated in the LDBC project. At the time the WIPP opened, 367 individuals had been measured using the *in vivo* protocol. This group of 367 measurements constitutes the pre-operational baseline to which subsequent results are compared. Counts performed after the opening of the WIPP are considered to be a part of the operational monitoring phase of the WIPP EM. Recounts began in July 1999, and 198 recount bioassays had been performed through 1 October 2002. In addition, 62 new volunteers have participated in the program since 1 October 2001.

Demographic characteristics (Table 26) of the current LDBC cohort are statistically unchanged from those reported in previous CEMRC reports, and are generally consistent with those reported in the 1990 census for citizens living in Carlsbad. The largest deviation between the LDBC cohort and 1990 census is under-sampling of Hispanics. Demographic characteristics of the LDBC project will be compared to results of the 2000 census in the near future, which will provide a more accurate representation of the current population. In addition, it is important to note that if the presence of a radionuclide is dependent on a subclass of interest (gender, ethnicity, etc.), valid population estimates can still be made by correcting for the proportion of under- or over-sampling for the particular subclass. It is important to note that operational monitoring includes the counting of new individuals and the recounting of previously measured participants. Based on the data reported herein, there is no evidence of an increase in the frequency of detection of internally deposited radionuclides for citizens living within the vicinity of the WIPP, since the WIPP began receipt of radioactive waste.

As discussed in detail in the CEMRC 1998 Report and elsewhere (Webb et al., 2000), the criterion, L_C, was used to evaluate whether a result exceeds background, and the use of this criterion will result in a statistically inherent 5% false-positive error rate per pair-wise comparison (5% of all measurements will be determined to be positive when there is no activity present in the person). For the baseline measurements (N = 367), the percentage of

results greater than L_C were consistent with a 5% random false-positive error rate, at the 95% confidence level (1 to 9%), for all radionuclides except ^{232}Th via the decay of ^{212}Pb, ^{235}U / ^{226}Ra, ^{60}Co, ^{137}Cs, ^{40}K, ^{54}Mn, ^{232}Th via the decay of ^{228}Ac (Table 27). As discussed in detail in the 1998 report, five of these (^{232}Th via ^{212}Pb, ^{60}Co, ^{40}K, ^{54}Mn (^{228}Ac interference) and ^{232}Th (via ^{228}Ac)) are part of the shield-room background and positive detection is expected at low frequency. ^{40}K is a naturally occurring isotope of an essential biological element, so detection in all individuals is expected. ^{137}Cs and ^{235}U / ^{226}Ra are not components of the shielded room background and were observed at frequencies greater than the 95% confidence interval for the false positive error rate (discussed in more detail later).

For the operational monitoring counts (Table 27, N = 465), the percentage of results greater than L_C were consistent with baseline at a 95% confidence level (margin of error), except for ^{60}Co and ^{232}Th (via ^{228}Ac). For these radionuclides, the percentage of results greater than L_C decreased relative to the baseline. This would be expected for ^{60}Co, since the radionuclide has a relatively short half life (5 years), and the content within the shield has decreased via decay by approximately 39% since the baseline phase of monitoring. The differences in ^{232}Th (via ^{228}Ac) results between the baseline and operational monitoring phase were observed last year and are likely due to the replacement of aluminum (tends to contain Th and U) in some of the detector cryostat components with those manufactured from low radiation background steel.

^{40}K results were positive for all participants and ranged from 1024 to 5557 Bq per person with an overall mean (± SE) of 2634 (± 30) Bq per person. Such results are expected since K is an essential biological element contained primarily in muscle, and a theoretical constant fraction of all naturally occurring K is the radioactive isotope ^{40}K. The mean ^{40}K value for males (± SE), was 3194 (± 33) Bq per person, which was significantly greater ($p < 0.0001$) than that of females, which was 1979 (± 25) Bq per person. This result was expected since, in general, males tend to have larger body sizes and greater muscle content than females.

Detectable ^{137}Cs is present in 25 ± 3% (95% confidence level, baseline and operational monitoring counts) of citizens living in the Carlsbad area. These results are consistent with findings previously reported in CEMRC reports and elsewhere (Webb et al., 2000). Detectable ^{137}Cs body burdens ranged from 4.9 to 63.1 Bq per person with an overall mean (± SE) of 10.1 (± 0.4) Bq per person. The mean ^{137}Cs body burden for males (± SE), was 10.8 (± 0.6) Bq per person, which was significantly greater ($p = 0.002$) than that of females, which was 8.7 (± 0.3) Bq per person. As previously reported (CERMC Reports; Webb et al., 2000) the presence of ^{137}Cs was independent of ethnicity, age, radiation work history, consumption of wild game, nuclear medical treatments and European travel. However, the occurrence of detectable ^{137}Cs was associated with gender where males had higher prevalence of ^{137}Cs relative to females. Furthermore, the presence of ^{137}Cs was associated with smoking (where smokers had a higher prevalence relative to non-smokers). It is likely that the association with gender is related to the tendency for larger muscle mass in males than in females, as supported by the ^{40}K results. The association of ^{137}Cs with smoking could be related to the presence of fallout ^{137}Cs in tobacco, decreased pulmonary clearing capability in smokers, or other as yet unidentified factors. Further study is warranted, but no additional investigations into this observation were conducted during this reporting period.

As reported in previous CEMRC reports, the percentage of results greater than L_C for ^{235}U / ^{226}Ra (10%) are significantly (although slightly) higher than the distribution-free confidence interval for a 5% random false-positive error rate (1 to 9%). These data are not nearly as compelling as those for ^{137}Cs, but the large sample size of the current cohort tends to support the observed pattern. Although ^{235}U and ^{226}Ra cannot be differentiated via gamma spectroscopy, it is likely the signal is the result of ^{226}Ra because the natural abundance of ^{226}Ra is much greater than that of ^{235}U. Although further study is needed; no additional investigations into this

observation were conducted during this reporting period.

Survey Results

An ongoing survey is being conducted for the following purposes: (a) to obtain a measure of the public's awareness of the Center's WIPP-EM program, (b) to gauge their perception of risks posed by operations at the WIPP site, and (c) to identify the environmental media that the public feels it is most important to monitor. As of September 2002, 158 people who have been counted under the Lie Down and Be Counted program have participated in the survey.

Seventy-four percent of the 158 respondents said they were aware that the Center had an environmental monitoring program beyond the LDBC program (Table 28). Of the 117 people who said they were aware of the Center's environmental monitoring program, less than half responded that they have seen at least some of the results from the program, with the majority of those reporting that they had seen results published in either the Center's newsletter or annual report. Fourteen percent of all the people surveyed said that they had logged-on to the Center's web site at least one time.

When asked if they were concerned that the WIPP site may affect community health or the environment 64% of the people polled responded "No." The 57 people who responded "Yes" had an average score of 2.8 when asked to rate their level of concern on a scale of 1 (not very concerned) to 5 (very concerned). Interestingly, these results appear to be at odds with the level of concern found in a larger survey (CEMRC, 1998) conducted between March 1995 and January 1996. In that prior study, between 33% to 42% of the people responded that they were "very concerned" about radioactive waste, and 41% to 49% of the people said they were "very concerned" about toxic waste while only 14% to 17% and 9% to 15% of the people said that were "not at all concerned" about radioactive or toxic waste, respectively. The results of the current survey may signal a shift in the local population's perception of the risk to community health and the environment posed by the WIPP site since it opened. However, one cannot draw definitive conclusions from this comparison because the sample size of the current survey is small and the results may be biased by the fact that the people polled in the current survey have been actively participating in the Center's LDBC program. {It is worth noting, however, that if the latter is the case, one could argue that the LDBC program has succeeded in assuaging public concerns over the WIPP.}

To gain some insight into the public's attitude about environmental monitoring and what they perceive as important pathways for contaminants, the persons who were surveyed were asked to rank from 1 (not very important) to 5 (very important), the importance of monitoring six specific environmental media, including air, drinking water, plants/animals, river/lake sediments, river/lake water and soils (Table 29). The average scores ranged from 4.5 (drinking water) down to 3.9 (plants/animals; river/lake sediments and soils) with drinking water and air ranking the highest (most important) and soils, river/lake sediments and plants/animals ranking the lowest (least important). However, with the lowest mean score of 3.9, the results suggest that the majority of the people feel that it was somewhat to very important to maintain some level of monitoring across all the categories presented. These results are consistent with the findings in the 1998 survey report in which the majority of the respondents indicated they were at least somewhat concerned with "pollution in the environment", "toxic waste", "radioactive waste" and "contaminants in water".

Table 26. Demographic Characteristics of the "Lie Down and Be Counted" Population Sample through 1 October 2002

Characteristic		2002 Sample Group ([a]margin of error)	[b]Census, 1990
Gender	Male	52.7% (48.7 to 56.6%)	48.0%
	Female	47.3% (43.4 to 51.3%)	52.0%
Ethnicity	Hispanic	14.2% (11.5 to 16.9%)	33.4%
	Non-Hispanic	82.5% (79.5 to 85.5%)	63.0%
	Other	3.3% (1.9 to 4.7%)	3.6%
Age 60 or older		30.8% (27.1 to 34.4%)	33.7%
Currently or previously classified as a radiation worker		5.8% (3.9 to 7.7%)	[c]NA
Consumption of wild game within last 3 months		18.8% (15.6 to 21.9%)	NA
Medical treatment, other than x-rays, using radionuclides		7.9% (5.7 to 10.1%)	NA
European travel within the last 2 years		3.9% (2.4 to 5.5%)	NA
Current smoker		14.5% (11.7 to 17.4%)	NA

[a]The margin of error represents the 95% confidence interval of the observed proportion.; under complete replication of this experiment, one would expect the confidence interval to include the true population proportion 95% of the time if the sample was representative of the true population.
[b]United States Department of Commerce, Economics and Statistics Administration, Bureau of the Census. *1990 Census of Population*. Washington, DC: US Government Printing Office
[c]NA = not available

Table 27. "Lie Down and Be Counted" Results through 1 October 2002

Radionuclide	*In Vivo* Count Type	Baseline Counts (prior to 27 March 1999) [a]N = 367	Operational Monitoring Counts (27 March 1999 – 1 October 2002) N = 465	
		% of Results ≥ [b]L_C	% of Results ≥ L_C	[c]Margin of Error (%)
^{241}Am	Lung	5	4	2 to 6
^{144}Ce	Lung	5	3	2 to 5
^{252}Cf	Lung	4	6	4 to 8
^{244}Cm	Lung	6	5	3 to 6
^{155}Eu	Lung	7	5	3 to 7
^{237}Np	Lung	4	3	2 to 5
^{210}Pb	Lung	4	6	3 to 8
Plutonium Isotope	Lung	6	5	3 to 8
[d] ^{232}Th via ^{212}Pb	Lung	34	36	31 to 40
^{232}Th	Lung	5	5	3 to 7
^{232}Th via ^{228}Th	Lung	4	5	3 to 7
^{233}U	Lung	6	9	6 to 12
^{235}U / ^{226}Ra	Lung	11	10	7 to 13
Natural Uranium via ^{234}Th	Lung	5	6	4 to 8
^{133}Ba	Whole Body	4	3	2 to 5
^{140}Ba	Whole Body	5	5	3 to 6
^{141}Ce	Whole Body	4	4	2 to 6
^{58}Co	Whole Body	4	3	2 to 5
[d] ^{60}Co	Whole Body	55	32	28 to 36
^{51}Cr	Whole Body	6	5	3 to 6
^{134}Cs	Whole Body	2	3	1 to 4
^{137}Cs	Whole Body	28	22	18 to 26
^{152}Eu	Whole Body	7	5	3 to 7
^{154}Eu	Whole Body	4	4	2 to 5
^{155}Eu	Whole Body	4	4	2 to 5
^{59}Fe	Whole Body	4	7	4 to 9
^{131}I	Whole Body	5	3	2 to 5
^{133}I	Whole Body	3	4	2 to 5
^{192}Ir	Whole Body	4	4	2 to 6
^{40}K	Whole Body	100	100	[e]NC
[d] ^{54}Mn	Whole Body	12	10	7 to 13
^{103}Ru	Whole Body	2	2	0 to 3
^{106}Ru	Whole Body	4	4	2 to 5

Table continued on next page

Table 27. "Lie Down and Be Counted" Results through 1 October 2001 (Cont.)

Radionuclide	*In Vivo* Count Type	Baseline Counts (prior to 27 March 1999) [a]N = 367	Operational Monitoring Counts (27 March 1999 – 1 October 2002) N = 465	
		% of Results ≥ [b]L_C	% of Results ≥ L_C	[c]Margin of Error (%)
^{125}Sb	Whole Body	5	4	2 to 6
^{232}Th via ^{228}Ac	Whole Body	35	25	21 to 28
^{88}Y	Whole Body	8	6	4 to 8
^{95}Zr	Whole Body	7	5	3 to 6

[a]N = number of individuals

[b]To determine whether or not activity has been detected in a particular person, the parameter L_C is used; the L_C represents the 95th percentile of a null distribution that results from the differences of repeated, pair-wise background measurements; an individual result is assumed to be statistically greater than background if it is greater than L_C

[c]The margin of error represents the 95% confidence interval of the observed percentage; under complete replication of this experiment, one would expect the confidence interval to include the true population percentage 95% of the time, if the sample was representative of the true population

[d]These radionuclides are present in the shield background, so they are expected to be detected periodically

[e]NC = not calculated; the margin of error for ^{40}K cannot be calculated since this radionuclide is present in all individual.

Table 28. Summary of Environmental Monitoring Telephone Survey Responses

Survey Question	[a]Summary of Answers	Comment
1. Were you aware of the Center's environmental monitoring program (other than Lie Down & Be Counted program)?	Yes – 74% (67% – 81%) No – 26% (19% – 33%)	Based on the total number of 158 respondents
2. Have you seen any of the results from the program?	Yes – 38% (29% - 46%) No – 62% (53% - 72%)	Based on the total number of 117 people who answered "Yes" to question 1.
2b. If "Yes" (to question 2), where have you seen these results?	Center Newsletter – 14 Center Annual Report – 11 Center Web Site – 6 Newspaper – 6 Center Posters – 2 Center Presentations – 3 Talking to Center Employees – 4 WIPP Publication – 1 Dept. Develop. Meeting – 1 City Council – 1 At WIPP – 1	Based on the 44 people that answered "Yes" to question #2
3. Have you ever visited the Center's web site?	Yes – 14% (8% - 20%) No – 86% (80% - 92%)	Based on the total number of 158 respondents
4. Are you concerned that the U.S. DOE WIPP site may affect this community's health and environment?	Yes – 36% (31% - 42%) No – 64% (56% - 71%)	Based on a total number of 157 respondents. (1 person did not want to answer the question)
4b. If "Yes" (to question 4), rate your concern on a scale of 1 to 5, where: 1 = Not Very Concerned 5 = Very Concerned	1 = 19% 2 = 26% 3 = 28% 4 = 8% 5 = 19% **Mean Score = 2.8**	Based on the 57 people that answered "Yes" to question #4

[a]Values in parentheses represent a 95% confidence interval

Table 29. Responses from 158 Individuals on the Importance of Monitoring Specific Environmental Media

[a]Score	Environmental Media					
	Air	**Drinking Water**	**Plant/Animals**	**River/Lake Sediments**	**River/Lake Water**	**Soils**
1 Not Very Important	5%	5%	8%	6%	6%	8%
2	3%	3%	4%	8%	5%	6%
3	9%	5%	27%	23%	15%	18%
4	14%	13%	16%	18%	22%	22%
5 Very Important	68%	73%	45%	44%	51%	46%
Mean Score	**4.4**	**4.5**	**3.9**	**3.9**	**4.1**	**3.9**

[a]Summed percent for some columns are less than 100% due to rounding error

APPENDICES

Appendix A. Brief History of Carlsbad Environmental Monitoring and Research Program

The Carlsbad Environmental Monitoring & Research Center (CEMRC) was created in 1991 as a division of the Waste-management Education & Research Consortium (WERC), in the College of Engineering at New Mexico State University (NMSU). The CEMRC was conceived as a result of inquiries to WERC by concerned citizens of the Carlsbad region, acting as a grassroots coalition who recognized the need for high quality, independent, health and environmental assessment data. Many individuals and organizations supported the CEMRC's formation including the residents of Carlsbad, NM, and the surrounding region; NMSU; the Carlsbad Department of Development; the New Mexico Congressional Delegation; the New Mexico Radioactive and Hazardous Materials Committee; Westinghouse Electric Corporation; and the U.S. Department of Energy (DOE). The CEMRC was established with a grant entitled "Carlsbad Environmental Monitoring and Research Program" (CEMRP) from DOE to NMSU. The CEMRP initially was funded for $27 million over a seven-year period (1991–1998). Subsequently, the grant was increased to almost $33 million to support operations of the program until 2008.

Dr. Rohinton (Ron) K. Bhada served as Project Director for the CEMRP during 1991-1999. Dr. Donald J. Fingleton served as Director of the CEMRC during 1991-1996. In 1996, Dr. Fingleton was named Director of Laboratory Development, and Dr. Marsha Conley became Director of Operations. Dr. Fingleton was transferred to a position with WERC in 1997, and Dr. Conley became Director. Mr. Joel Webb was named Manager of Program Development in 1998. Dr. Conley was named CEMRP Project Director in 1999. In July 2001, Dr. Conley retired. In February 2002, Mr. Joel Webb was appointed Director of CEMRC and CEMRC Project Director. Dr. George Hidy acted as an interim director during the search for the new permanent director.

Temporary office accommodations for the CEMRC initially were provided at NMSU-Carlsbad. In 1992, the CEMRC moved to a leased facility at 800 West Pierce in Carlsbad, which served as a basis for operations through December 1996. Flatow Moore Bryan Shaffer McCabe Architects (Albuquerque, New Mexico) and Research Facilities Design (San Diego, California) were selected in 1991 to design the CEMRC's new facilities. In December of 1993, DOE Secretary Hazel O'Leary made a commitment to provide approximately $7 million in additional funding to support debt service for construction of the new facility. In 1994, the NMSU Board of Regents approved the sale of New Mexico State University Research Corporation Lease Revenue bonds to secure construction money. Construction of the Phase I facility began in August 1995 and was completed in December 1996. The facility is located adjacent to the NMSU-Carlsbad campus, on 22 acres of land donated to NMSU by then New Mexico State Representative Robert S. Light (D-55th District). On March 23, 1997, the Phase I facility was named the Joanna and Robert Light Hall (to be referred to as Light Hall).

In addition to work associated with design and construction of buildings for the CEMRC, a variety of other developmental projects were undertaken to support the CEMRC's scientific activities. In 1993, design began for the Mobile Bioassay Laboratory (MBL) that would complement the facilities planned for the new CEMRC building. Construction of the MBL began in 1994, and the unit was completed and delivered to Carlsbad in 1996. An application for a Radioactive Material License was prepared and submitted to the New Mexico Environment Department, and the license was issued in 1996.

In 1999, CEMRC was separated from WERC and is now a division reporting directly to the Dean of Engineering at NMSU.

Appendix B. Subcontracts for Technical Assistance during 2002

Subcontractor	Scope of Work
Perini Environmental, L.L.C	Sample Collection for Characterization of Radioactive Elements in Oil and Gas Production in the Vicinity of the DOE Waste Isolation Pilot Plant
[a] University of Texas at El Paso	Investigations of the Low-Wind Particulate Matter Spikes at the NMED Sunland Park City Yard Monitoring Site
National Institute of Standards & Technology	Intercomparison services for radioanalyses

[a] Collaborative work not funded through CEMRC

Appendix C. Members of Scientific Advisory Board (SAB) and Program Review Board (PRB)

Member/Term of Service	Affiliation
John M. Briggs, Ph.D. (SAB) / 2000-present	Associate Professor, Department of Plant Biology, Arizona State University, Tempe, Arizona
Paul M. Bertsch, Ph.D. (SAB) / 2000-present	Director, Savannah River Ecology Laboratory, University of Georgia, Aiken, South Carolina
Judith Chow, Ph.D. (SAB) / 2000-present	Research Professor, Desert Research Institute, Reno, Nevada
George M. Hidy, D.Eng. (PRB) / 2000-present	Consultant, Envair/Aerochem President (past), Desert Research Institute, Reno, Nevada
Shawki A. Ibrahim, Ph.D. (SAB) / 2000-present	Professor, Department of Radiological Health Sciences, Colorado State University, Ft. Collins, Colorado
Gary H. Kramer, Ph.D. (SAB) / 2000-present	Head, Human Monitoring Laboratory, Radiation Protection Bureau, Health Canada, Ottawa, Ontario, Canada
Roger McClellan, Ph.D. (PRB)/2001-present	Consultant; President Emeritus, Chemical Industry Institute of Toxicology, Albuquerque, New Mexico
David E. Reichle, Ph.D. (PRB) / 2000-present	Associate Director Emeritus, Life Sciences and Environmental Technologies, Oak Ridge National Laboratory, Oak Ridge, Tennessee

Appendix D. Professional Presentations and Publications during 2002

Author	Title	Publisher/Conference
Savoie, D.L., **R. Arimoto**, W.C. Keene, J.M. Prospero, R.A. Duce, and J.N. Galloway	Marine biogenic and anthropogenic contributions to non-sea-salt sulfate in the marine boundary layer over the North Atlantic Ocean	Journal of Geophysical Research, Vol. 107, No. D18, 3:1-21
Arimoto, R., S. Sage, C. Schloesslin, T. Kirchner, J. Webb, B. Stewart, D. Schoep, and **M. Walthall**	Abstract: Man and dust: a unique perspective from southeastern New Mexico/west Texas	In: Lee, J. A. and T. M. Zobek, Proceedings of ICAR5/GCTE-SEN Joint Conference, Texas Tech University, Lubbock, TX, USA Publication 02-2: 148-151, 2002
Arimoto, R., T. Kirchner, J. Webb, B. Stewart, D. Schoep, M. Walthall	Abstract: "Chemical and radiochemical studies of aerosol particles from the vicinity of the WIPP"	Book of Abstracts, American Chemical Society, 17th Rocky Mountain Regional Meeting, p. 37, 2002
Arimoto, R., W. Balsam, and **C. Schloesslin**	Visible spectroscopy of aerosol particles collected on filters: Iron-oxide minerals	Atmospheric Environment, 36:89-96, 2002
Kirchner, T.B., J.L. Webb, S.B. Webb, **R. Arimoto, D. Schoep,** and **B.D. Stewart**	Variability in background levels of surface soil radionuclides in the vicinity of the Waste Isolation Pilot Plant	Journal of Environmental Radioactivity, 60:275-291, 2002
Whicker, J.J., D.D. Breshears, P.T. Wasiolek, **T.B. Kirchner**, R.A. Tavini, **D.A. Schoep**, J.C. Rodgers	Temporal and spatial variation of episodic wind erosion in unburned and burned semiarid shrubland	Journal of Environmental Quality, 31(2):599-612, 2002
Zhang, X.Y., J.J. Cao, L.M. Li, **R. Arimoto**, Y. Cheng, B. Huebert, and D. Wang	Characterization of atmospheric aerosol over XiAn in the south margin of the Loess Plateau, China	Atmospheric Environment, 36:4189-4199, 2002
Arimoto, R.	Major ions and tract elements in aerosols from the South Pole	American Geophysical Union, Fall Meeting, San Francisco, CA (Poster), December 2001.
Arimoto, R.	Desert Dust	Chihuahuan Desert Conservation Alliance, Carlsbad, NM, January 2002.
Arimoto, R.	Man and aerosol biogeochemical cycles in the anthropocene	University of North Texas, Denton, Texas, March 2002.
Arimoto, R.	Aerosol composition in Korea and China during ACE-Asia.	ACE-Asia Data Workshop, Beijing, People's Republic of China, April 2002.
Arimoto, R.	Eolian dust, chemistry, and climate	University of Texas, Arlington, April 2002.
Arimoto, R.	Man and dust: a unique perspective from southeastern New Mexico/West Texas	International Conference on Aeolian Research and The Global Change & Terrestrial Ecosystem-Soil Erosion Network Joint Conference, Texas Tech University, Lubbock TX, July 2002.

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Appendix D. Professional Presentations and Publications during 2002 (cont.)

Author	Title	Publisher/Conference
Arimoto, R.	Asian dust, chemistry and climate	Sixth International Aerosol Conference, Taipai, Taiwan, September 2002.
Arimoto, R.	Chemical and radiochemical studies of aerosol particles from the vicinity of the WIPP	American Chemical Society, 17th Rocky Mountain Regional Meeting, Albuquerque, NM, October 2002.
Arimoto, R.	Asian dust storms: some interlocking questions, issues and unknowns	2002 International Workshop of Dust Storm, Beijing, People's Republic of China, October 2002.
Arimoto, R.	Dry Deposition of Trace Elements to the Western North Atlantic	American Geophysical Union, Fall Meeting, San Francisco, CA (Poster), December 2002.

Appendix E. Guest Colloquia

Topic	Presenter
New Insight in Third Phase Formation in the TBP–Alkane System	Dr. Marian Borkowski, Deputy Head, Radiochemistry Department, Institute of Nuclear Chemistry and Technology, Warsaw, Poland and Visiting Scientist, Chemistry Division, Argonne National Laboratory
The Management of Radioactive Waste in Hungary	Dr. Tamas Hamor, a national expert of the Joint Research Centre of the European Commission, Institute for Environment and Sustainability, Soil and Waste Unit
Synchrotron XAS applications in actinide molecular environmental science: Reduction of pertechnetate and Pu(VI) by metallic Fe and Al	Mei Ding, Ph.D.**,** Los Alamos National Laboratory, Chemistry Division
Chemical Speciation of Heterogeneously Reduced Plutonium Precipitated from Brines	Steven Conradson, Ph.D., Los Alamos National Laboratory, Chemistry Division
Effect of Water Alpha Radiolysis On the Nuclear Spent Fuel UO_2 Matrix Alteration	Dr. Jean-Francois Lucchini**,** Post-Doctoral Research Associate**,** Los Alamos National Laboratory
Stability and Reactivity of U(VI), Np(VI) and Pu(VI) in Brine	Dr. Donald T. Reed

Appendix F. Major Tours, Public Presentations, Exhibits and Other Outreach

Group/Activity
Sunset Elementary School student science fair - exhibit and learning activities
The Pate Elementary School fifth grade challenge class – tour and presentation
Carlsbad Christian Home Educators students – tour and presentation
Community Health and Safety Fair 2002 – exhibit
Relay for Life staff volunteer participation
River Blitz staff volunteer participation
Earth Day 2002 (Living Desert State Park) - exhibit
Ninth grade students from the New Mexico Academy, Santa Fe, New Mexico – tour and presentation
Leadership Carlsbad – tour and presentation
Agricultural Science Center Field Day 2002 - exhibit
CEMRC's lung and whole body counter was featured in the July issue of National Geographic
Project Playground staff volunteer participation

Appendix G. Leadership Participation by CEMRC Staff in Professional Functions

Function	CEMRC Staff/Role
[a]ISTC Project #1908 "Experimental and Theoretical Investigation of Dynamics and Kinetics of Gas Contamination and Aerosols in the Central Continental Asia (Baikal Region and Sayan Background Station – Mondy included)".	Richard Arimoto – "Foreign Collaborator"

[a]The International Science and Technology Center (ISTC) promotes the nonproliferation of weapons technology of mass destruction. Valery Anatolyevich Zagaynov, of the Karpov Institute of Physical Chemistry, Russia is the lead researcher.

Appendix H. New Project Development

Proposal/Bid Title	PI(s)	Sponsor	Funding/ Term	Status
An investigation of sulfur chemistry in the Antarctic troposphere	R. Arimoto (with Georgia Institute of Technology and others)	National Science Foundation	$160,000, 1998-2002	Funded, in progress
Collaborative research: aerosol characterization experiment (ACE)-Asia surface network implementation, operations, and coordination	R. Arimoto (with University of Virginia and others)	National Science Foundation	$139,968, 2000-2004	Funded, in progress
Collaborative research: integrated studies of morphological, chemical optical, and radiative properties of multi-component aerosols containing mineral dust in the ACE-Asia region	R. Arimoto (with University of Hawaii and others)	National Science Foundation	$148,923, 2000-2004	Funded, in progress
Air deposition of mercury and other airborne pollutants in an arid environment	R. Arimoto (with NMSU Cooperative Fish and Wildlife Research Unit and others)	Southwest Center for Environmental Research and Policy (with funding from U.S. Environmental Protection Agency)	$12,537, 2000-2002	Completed
Collaborative research: on Iron-oxide Minerals in Asian Dust	R. Arimoto	National Science Foundation	$64,207	Submitted, not funded
Investigation of the Low-Wind Particulate Matter Spikes at the NMED Sunland Park City Yard Monitoring Site	R. Arimoto	SCERP	$81,657, 2002-2003	Funded, in progress

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Appendix H. New Project Development (cont.)

Proposal/Bid Title	PI(s)	Sponsor	Funding/ Term	Status
Technology for Rapid Assessment of Long-Term Risk Management at Arid DOE Sites; Factoring in Land-Surface Dynamics, Soil Profile Changes, and Disturbance Events	T. Kirchner, M. Walthall	U.S. Department of Energy	$537,167, 2002-2005	Submitted, not funded
Limnological monitoring: Brantley Dam Reservoir	M. Walthall	U.S. Department of Interior, Bureau of Reclamation	$83,363, 1997-2003	Amended, in progress
Monitoring Soil Salinity, Water Quality and Sediment Load in and along the Pecos River during Salt Cedar Control Efforts	M. Walthall	Carlsbad Soil & Water Conservation District	$45,519, 2002-2004	Funded, in progress
Determination of Uranium and Other Elements by ICP-MS	D. Moir	Sandia National Laboratories	$20,000, 2002-2003	Funded, in progress
Collaborative research: CEMRC Support of INEEL Environmental Surveillance, Education, and Research	D. Moir	S. M. Stoller Corporation	$1,800, 2002	Completed
Determination of Beryllium Content in Solutions and Filters	D. Moir	Johns Hopkins University	$16,300, 2002-2003	Funded, in progress
Lung & whole body *in vivo* radiobioassay measurements	J. Webb (with David Schoep)	Waste Control Specialists, Inc.	$233,414, 1997-2001	Amended, in progress
In vivo radiobioassay measurements for WIPP personnel	J. Webb (with David Schoep)	Westinghouse Electric Company	$299,000, 1998-2002	Completed
Center for nuclear, neutrino and astroparticle physics	J. Webb (with Ohio State University and others)	National Science Foundation	$229,344, 2001-2006	Collaboration commitment re-submitted
Analytical scientific support for the Los Alamos National Laboratory, Carlsbad Office, actinide chemistry and repository science program	J. Webb (with B. Stewart, R. Arimoto and M. Walthall)	Los Alamos National Laboratory	$810,080, 2001-2002	Funded, in progress

Appendix I. Status of Completion of 2001 Key Performance Indicators

Focus Area	Key Performance Indicator	Status
Aerosol particles	Continue concurrent high-volume and low-volume/dichotomous sampling at current four locations through 2002.	**Ongoing.**
Soils	Review sampling program for redesign to facilitate efficient monitoring studies.	**Completed; rescheduled sampling for every other year, alternating with sediments and surface water.**
Meteorology	Continue concurrent operation of sampling stations at two current sites through 2002.	**Ongoing.**
Drinking water	Review sampling program for redesign to facilitate efficient monitoring studies.	**Completed; sampling is to continue to be performed annually.**
Sediment and surface water	Review sampling program for redesign to facilitate efficient monitoring studies	**Completed; rescheduled sampling for every other year, alternating with soils.**
Human studies	Complete repeat counts for original volunteer cohort, and initial counts for a minimum of 100 new volunteers	**The entire original cohort has been contacted at least once in an attempt to schedule recounts. The effort is on-going with some of the original cohort being scheduled for a third count. Several hundred people were contacted in 2002 through various functions in an effort to recruit new volunteers to the program. A total of 62 new volunteers were successfully recruited in 2002.**
Radiological analyses	Complete analyses of soil, aerosol, sediment, surface water and drinking water samples (collected through June 2002) by October 2002	**Not applicable for soils and surface waters and sediments. Completed for aerosols and drinking water.**
	Continue FAS sample analyses to meet weekly and quarterly posting schedule.	**Completed and ongoing.**
Non-radiological analyses	Complete analyses of representative subset of 2002 low-volume aerosol particles, soil, sediment, surface water and drinking water samples within three months after each sample collection	**Completed analyses of drinking water samples, which were the only non-aerosol samples collected for the WIPP EM. Analyses of the low-volume aerosol samples were completed with some minor delays due to changes in priorities. The low-volume aerosol analyses are now up-to-date.**
	Continue FAS sample analyses to meet weekly and quarterly posting schedule	**Completed.**

Table continued on next page

Appendix I. Status of Completion of 2001 Key Performance Indicators (cont.)

Focus Area	Key Performance Indicator	Status
Data management and dissemination	Post results of radioanalyses of 2002 and pre-2002 samples within two months after completion of analyses of each set of samples	**Not applicable for soils, surface waters and sediments. Ongoing for FAS, aerosols and drinking water.**
	Post results of non-radiological analyses of 2002 samples within two months after completion of analyses of each set of samples	**Not applicable for soils, surface waters and sediments. Ongoing for FAS, aerosols and drinking water.**
	Issue CEMRC 2001 Report; post report and background data to CEMRC web site by May 2002 (web site currently under development)	**2001 Report posted; background data not posted.**

REFERENCES

American National Standards Institute (ANSI). 1996. *Performance Criteria for Radiobioassay.* ANSI-N13.30.

American Public Health Association. 1981. *Standard Methods for the Examination of Water and Wastewater, 15th Edition.*

Arimoto, R., T. Kirchner, J. Webb, M. Conley, B. Stewart, D. Schoep, and M. Walthall. 2002. *^{239,240}Pu and inorganic substances in aerosols from the vicinity of the Waste Isolation Pilot Plant: The importance of resuspension.* Health Physics, 83:456.

Barisic, D., A. Vertacnik, and S. Lulic. 1999. *Cesium contamination and vertical distribution in undisturbed soils in Croatia.* J. Environ. Radioactivity, 46:361-374.

Carlsbad Environmental Monitoring & Research Center. 1999. *1999 Report*.

Carlsbad Environmental Monitoring & Research Center. 2000. *2000 Report*.

Carlsbad Environmental Monitoring & Research Center. 2001. *2001 Report*.

Carlsbad Environmental Monitoring & Research Center. 1998. *Survey of Factors Related to Radiation Exposure and Perceptions of Environmental Risks in Carlsbad, Loving, Malaga and Hobbs, New Mexico*

Chugg, J., G. Anderson, D. King, and L. Jones. 1971. *Soil Survey of Eddy County*, New Mexico, U. S. Dept. of Agriculture Soil Conservation Service, U.S. Government Printing Office, Washington, D.C.

City of Carlsbad Municipal Water System. 2002. *2002 Annual Consumer Report on the Quality of Your Drinking Water.*

City of Carlsbad Municipal Water System. 2001. *2001 Annual Consumer Report on the Quality of Your Drinking Water.*

The City of Carlsbad Municipal Water System. 2000. *2000 Annual Consumer Report on the Quality of Drinking Water.*

The City of Carlsbad Municipal Water System. 1999. *Annual Consumer Report on the Quality of Tap Water.*

The City of Carlsbad Municipal Water System. 1998. *Annual Consumer Report on the Quality of Tap Water.*

Collins, A., D. Walling, H. Sichingabula, and G. Leeks. 2001. *Using ^{137}Cs measurements to quantify soil erosion and redistribution rates for areas under different land use in the Upper Kaleya River basin, southern Zambia.* Geoderma, 104:299-323.

Coppinger, K., W. Reiners, I. Burke, and R. Olson. 1991. *Net erosion on a sagebrush steppe landscape as determined by Cs-137 distribution*, Soil Sci. Soc. Am. J., 55:254-258.

Cothern, C.R. and W.L. Lappenbusch. 1983. *Occurrence of uranium in drinking water in the US.* Health Physics, 45:89-99.

Dutkiewicz, V. A., and L. Husain. 1985. *Stratospheric and Tropospheric Components of Be in Surface Air*. Journal of Geophysical Research , 90:5783-5788.

Eisenbud, M. and T. Gesell. 1997. *Environmental Radioactivity from Natural, Industrial, and Military Sources*. Academic Press, San Diego.

Faller, S. 1994. *Residual soil radioactivity at the Gnome Test Site in Eddy County, New Mexico*, Report No. EPA 600/R-94/117, July 1994. Washington, D.C., Environmental Protection Agency.

Faller, S. 1994. *Gnome/Coach site disposal options*. U. S. Atomic Energy Commission NVO-131, 1973.

Gee, G.W. and J.W. Bauder. 1986. *Particle-Size Analysis.* In A. Klute. Methods of Soil Analysis: Part 1 – Physical and Mineralogical Methods, Agronomy Monograph no. 9 (2nd Edition), American Society of Agronomy – Soil Science Society of America, Madison, WI.

Graustein, W. and K. Turekian. 1996. *^{7}Be and ^{210}Pb indicate an upper troposphere source for elevated ozone in the summertime subtropical free troposphere of the eastern North Atlantic*. Geophysical Research Letters, 23, No. 5:539-542.

Isaksson, M., B. Erlandsson, and S. Mattsson. 2000. *A 10-year study of the ^{137}Cs distribution in soil and a comparison of Cs soil inventory with precipitation-determined deposition*, J. of Environ. Radioactivity, 55:47-59.

Kenney, J.W. 1994. *Preoperational Radiation Surveillance of the WIPP Project by EEG during 1992.* EEG-54.

Kenney, J. W., P. S. Downes, D. H. Gray, S. C. Ballard, Environmental Evaluation Group. 1995. *Radionuclide Baseline in Soil near Project Gnome and the Waste Isolation Pilot Plant.* EEG-58.

Kirchner, T.B., J. L. Webb, S. B. Webb, R. Arimoto, D. A. Schoep, and B. D. Stewart. 2002. *Variability in background levels of surface soil radionuclides in the vicinity of the Waste Isolation Pilot Plant.* Journal of Environmental Radioactivity 60:275-291.

Koch, D., D. Jacob, and W. Graustein. 1996. *Vertical transport of tropospheric aerosols as indicated by ^{7}Be and ^{210}Pb in a chemical tracer model*. Journal of Geophysical Research 101:18651-18666.

Krey, P., M. Heit, and K. Miller. 1990. *Radioactive fallout reconstruction from contemporary measurements of reservoir sediments*. Health Physics 59: 541-554.

Lee, S. C., K. A. Orlandini, J. Webb, D. Schoep, T. Kirchner, and D. J. Fingleton. 1998. *Measurement of baseline atmospheric plutonium-239,240 and americium -241 in the vicinity of the Waste Isolation Pilot Plant.* Journal of Radioanalytical Nuclear Chemistry, 234: 267-272.

Luo, S., T. Ku, R. Roback, M. Murrell, and T. McLing. 2000. *In-situ radionuclide transport and preferential groundwater flows at INEEL (Idaho): Decay-series disequilibrium studies*. Geochim. Cosmochim., 64 No. 5:867-881.

McAninch, J., T. Hamilton, T. Brown, T. Jokela, J. Knezovich, T. Ognibene, I. Proctor, M. Roberts, E. Sideras-Haddad, J. Southon and J. Vogel. 2000. *Plutonium measurements by accelerator mass spectrometry (AMS) at LLNL*. Nuclear Instruments & Methods in Physics Research Section B-Beam Interactions with Materials and Atoms 172: 711-716.

Mitchell, P., L. Vintró, H. Dahlgaard, C. Gascó and J. A. Sánchez-Cabeza. 1997. *Purturbations in the $^{240}Pu/^{239}Pu$ global fallout ratio in local sediments following nuclear accidents at Thule (Greenland) and Palomares (Spain)*. Science of the Total Environment 202: 147-153.

Otis Water User Co-Op. 1999. *Annual Water Quality Report*.

Owens, P., D. Walling, and Q. He. 1996. *The behavior of bomb-derived Cesium-137 fallout in catchment soils*, J. Environ. Radioactivity, 32:169-191.

Perkins, R.W. and C.W. Thomas. 1980. *Worldwide Fallout.* In: Hanson W. C., ed. Transuranic Elements in the Environment. U.S. Department of Energy: DOE/TIC-22800, 53-82.

Pimentel, D., C. Harvey, P. Resosudarmo, K. Sinclair, D. Kurz, M. McNair, S. Crist, L. Shpritz, L. Fitton, R. Saffouri, and R. Blair. 1995. *Environmental and economic costs of soil erosion and conservation benefits*. Science 267:1117-1123.

Prospero, J. M. and R.T. Nees. 1986. *Impact of the North African drought and El Niño on mineral dust in the Barbados trade winds.* Nature 320:735-738.

Prospero J. and R. Nees. 1977. *Dust concentration of the equatorial North Atlantic: Possible relationship to the Sahelian Drought*. Science, 196:1196-1198.

Ritchie, J.C., and J.R. McHenry. 1990. *Application of radioactive fallout cesium-137 for measuring soil erosion and sediment accumulation rates and patterns: a review*, J. Environ. Qual., 19:215-233.

Schimmack, W., K. Auerswald, and K. Bunzl. 2002. *Estimation of soil erosion and deposition rates at an agricultural site in Bavaria, Germany, as derived from fallout radiocesium and plutonium as tracers,* Naturwissenschaften, 89:43-46.

Staunton, S. and P. Levacic. 1999. *Cs adsorption on the clay-sized fraction of various soils: effect of organic matter destruction and charge compensating cation*, J. Environ. Radioactivity, 45:161-172.

Taylor, S.R. and S. M. McLennan. 1995. *The geochemical evolution of the continental crust.* Rev. Geophys., 33: 241-265.

U.S. Department of Energy. 1998. *Department of Energy Laboratory Accreditation Program for Radiobioassay.* DOE-STD-1112-98, Washington, DC.

U.S. Environmental Protection Agency. 1997. *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods.* EPA/SW-846.

U. S. Environmental Protection Agency. 1994. *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*. EPA/SW-846.

U.S. Environmental Protection Agency. 1994. *U.S. EPA Contract Laboratory Program National Functional Guidelines for Inorganic Data Review*. EPA 540/R-94013.

U.S. Environmental Protection Agency. 1983. *Methods for Chemical Analysis of Water and Wastes.* EPA 600/4-79-020.

U.S. Salinity Laboratory Staff. 1961. Diagnosis and improvement of saline and alkali soils. U.S. Department of Agriculture Handbook No. 60. U.S. Govt. Printing Office, Washington, D.C.

Vintró, L., P. Mitchell, O. Condren, A. Downes, C. Papucci and R. Delfanti. 1999. *Vertical and horizontal fluxes of plutonium and americium in the western Mediterranean and the Strait of Gibraltar*. Science of the Total Environment, 237/238:77-91.

Walling, D.E. and T.A. Quine. 1991. *Use of Cs-137 measurements to investigate soil erosion on arable fields in the UK: potential applications and limitations*, Journal of Soil Science, 42:147-165.

Webb, J.L. and T.B. Kirchner. 2000. *An evaluation of in vivo sensitivity via public monitoring.* Radiation Protection Dosimetry, 89 (3-4): 183-191.

Whicker, J., D. Breshears, P. Wasiolek, T. Kirchner, R. Tavani, D. Schoep, and J. Rodgers. 2002. *Temporal and spatial variation of episodic wind erosion in unburned and burned semiarid shrubland*, J. Environ. Qual., 31:599-612.

Winkler, R., F. Dietl, G. Frank, and J. Tschiersch. 1998. Temporal variation of ^{7}Be and ^{210}Pb size distributions in ambient aerosol. Atmospheric Environment 32, 983.

Waste Isolation Pilot Plant. 2000. *Confirmatory Volatile Organic Compound Monitoring Plan*". WP 12-VC.01 Revision 3.

Waste Isolation Pilot Plant. 2001. *Quality Assurance Project Plan for Confirmatory Volatile Organic Compound Monitoring*". WP 12-VC.02, Revision 4.