

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

*College of Engineering  
New Mexico State University*

Issued February 1, 2000



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## Acronyms and Abbreviations

μBq	microbecquerel
μm	micrometer
AA, AAS	atomic absorption, atomic absorption spectrometry
Am	americium
BOMAB	bottle mannequin absorption phantom
BTEX	ethylbenzene, m,p-xylene, o-xylene, toluene
C	centigrade
Cd	cadmium
Ce	cerium
CEMRP	Carlsbad Environmental Monitoring and Research Program
CEMRC	Carlsbad Environmental Monitoring & Research Center
CFR	Code of Federal Regulations
cm	centimeter
Cm	curium
Co	cobalt
Cs	cesium
Cu	copper
CV	coefficient of variation
DES	Duke Engineering and Services
df	degrees of freedom
DL	detection limit
DOE	U.S. Department of Energy
DRI	Desert Research Institute
EM	Environmental Monitoring
EML	Environmental Monitoring Laboratory
EPA	U.S. Environmental Protection Agency
Eu	europium
eV	electron volts
Fe	iron
g	gram
Ge	germanium
HCl	hydrochloric acid
HF	hydrofluoric acid
HNO <sub>3</sub>	nitric acid
hr	hour
IC	ion chromatography
ICP-ES	inductively coupled plasma emission spectrometry
ICP-MS	inductively coupled plasma mass spectrometry
K	potassium
keV	kiloelectron volts
km	kilometer
L	liter
m	meter
mb	millibar
MBL	mobile bioassay laboratory
mBq	millibecquerel



MCL	maximum contaminant level
MDA	minimum detectable amount
MDC	minimum detectable concentration
min	minute
MJ	mega joules
mL	milliliter
mm	millimeter
NEI	Nuclear Energy Institute (originally the Atomic Industrial Forum)
NIST	National Institute of Standards and Technology
NMAC	New Mexico Administrative Code
NMSU	New Mexico State University
Np	neptunium
Pb	lead
pH	scale indicating acidity or alkalinity of a substance
PM <sub>10</sub>	particulate matter smaller than 10 micrometers in diameter
PM <sub>2.5</sub>	particulate matter smaller than 2.5 micrometers in diameter
PRB	Program Review Board
Pu	plutonium
QA	quality assurance
QAP	quality assurance program
QC	quality control
RIP	Radiochemistry Intercomparison Program
ROI	region of interest
Ru	ruthenium
Rn	radon
Sb	antimony
SAB	Science Advisory Board
SD	standard deviation
SE	standard error
sec	second
shield	cast iron enclosure
SDWA	Safe Drinking Water Act
SWAT	Soil Water Air Testing
T <sub>1/2</sub>	half-life
Th	thorium
TPH	total petroleum hydrocarbons
TSP	total suspended particulate
U	uranium
UVB	Ultra-Violet B
W	watt
WERC	Waste-management Education & Research Consortium
WIPP	Waste Isolation Pilot Plant
XRF	X-ray fluorescence





Preparation of aerosol samples



Secretary of Energy, Bill Richardson at CEMRC exhibit



Aerosol Sampler at South Pole Station

**FORWARD**

*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>.*

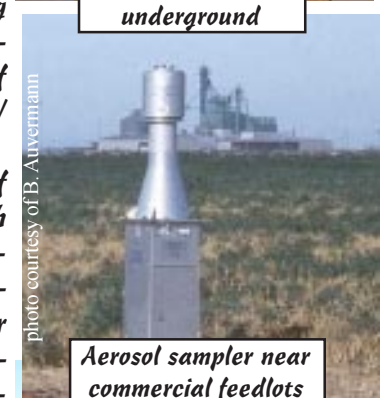


Map for volunteer project



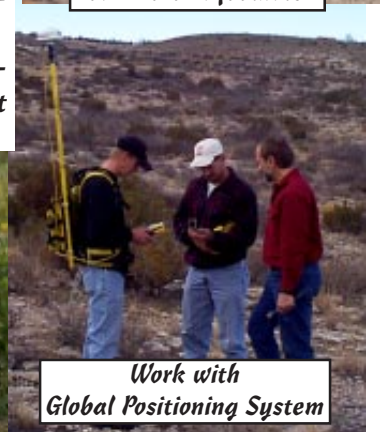
In vivo bioassay experiment in WIPP underground

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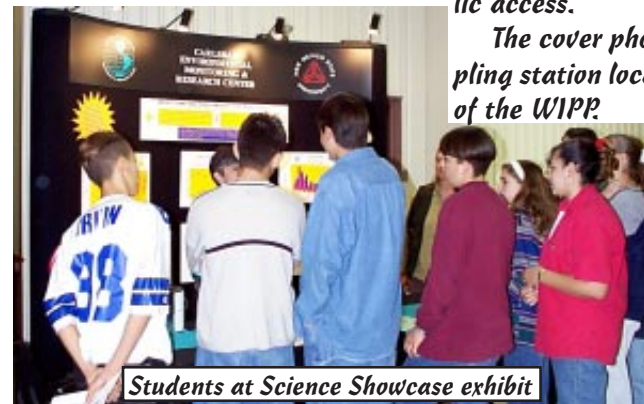


Aerosol sampler near commercial feedlots

*The cover photograph is the CEMRC aerosol sampling station located approximately 19 km southeast of the WIPP.*



Work with Global Positioning System



Students at Science Showcase exhibit



Desert in bloom

photo courtesy of B. Auvermann



## **OVERVIEW**

## Current Program Status

### History and Focus

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

- Establish a permanent center of excellence to anticipate and respond to emerging health and environmental needs
- 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 include 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 funding and service contracts. The CEMRC's primary objectives are to:

- Provide for objective, independent health and environmental monitoring
- Provide advanced training and educational opportunities
- Develop improved measurement methods, procedures and sensors
- 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 1998, staffing reached 27 professional and classified employees. At the end of 1999, the CEMRC employed 28 personnel (Table 1). Two scientific positions are open and under 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<sup>2</sup> laboratory and office facility constructed adjacent to the NMSU-Carlsbad campus. The CEMRC's scientific activities are organized into five 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) field operations, (2) internal dosimetry, (3) informatics and modeling, (4) radiochemistry and (5) environmental chemistry. Detailed descriptions of each program area and associated facilities and instrumentation are presented on the CEMRC's web site at <http://www.cemrc.org>.

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

CEMRC has begun participation with the DOE/Carlsbad Area Office (CAO) in studies to quantify the natural radiation background in the WIPP underground. The results of the studies are being used by CAO in efforts to

establish a Center for Applied Repository and Underground Science and recruit external research groups who could make use of the WIPP for projects that require low-radiation environments. These efforts could lead to new collaborations with scientists in a variety of disciplines, including astrophysics, health physics and genetics.

Program needs for external laboratory services have declined, but a few sub-contractual agreements were maintained or initiated to provide specific advanced methodologies for selected analyses (Appendix B). In addition to services provided by external organizations, several NMSU departments and divisions also provided support to the CEMRC for specific projects, including the Soil, Air and Water Testing Laboratory (SWAT), the Waste-management Education and Research Consortium (WERC) and the Fishery and Wildlife Science Department. With respect to collaborative research, 17 of the 1999 publications and presentations by CEMRC staff were co-authored with external colleagues, and eight of the CEMRC's proposed and existing new 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). Each SAB member visited the CEMRC during 1999 to review the individual program areas and provide expert guidance and consultation to the program leaders. Each program leader used the SAB observations and recommendations in structuring specific developmental goals, new experiments and methods improvements. Program leaders provided SAB members with follow-up reports prior to each SAB member's visit during 1999. The term of service for SAB members is two years, and new members for 2000-2001 terms have been identified (Appendix C).

The Program Review Board (PRB) for the CEMRC consists 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. Members of the PRB visited the CEMRC as a group during 1999, reviewed the overall operation of the CEMRC, and provided a joint report to the administration. An action plan responding to the review was prepared by the CEMRC director, and implementation of the plan by the director and NMSU administration is in progress. A follow-up report will be provided to the PRB members prior to their visit during 2000.

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

Current administrative staff includes a director, a fiscal specialist, a buyer specialist, a project manager, a manager of program development, a quality assurance manager and an administrative secretary. During 1999, partial support was also provided for three WERC administrators at NMSU, to assist in coordination with main campus business and with the WERC educational and research programs. Cumulative funding from the DOE for the CEMRP totaled approximately \$20.4 million through 1 October 1999, including \$0.5 million de-obligated from committed funds in March 1999. Cumulative expenditures by the CEMRP for the same period totaled approximately \$20.1 million. Proposed new funding for the 2000 Federal fiscal year is approximately \$3.4 million. Combined with carry-over funds, the projected CEMRP 2000 budget is approximately \$3.75 million.

Formal tracking of CEMRP project schedules and deadlines 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 accountabilities and track progress. Administrative and individual program area staff also have regularly scheduled review and planning sessions. Significant accomplishments and events are reported in monthly summaries provided to the DOE, NMSU, SAB and PRB.

## **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 14 presentations at international, national and regional scientific meetings and 20 papers were published, are in press, or have been submitted for publication in peer-reviewed scientific journals and books during 1999 (Appendix D). A cumulative list of publications by CEMRC staff since 1996 is presented on the CEMRC web page.

The CEMRC issued a 1998 report that presented extensive data on radionuclides, non-radioactive constituents and other basic environmental parameters from the WIPP Environmental Monitoring project. These reports 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>. The CEMRC web site was featured in the September 1, 1999 issue of *Environmental Science & Technology* (Appendix D).

A notable new feature, the "FAS Lane", was added to the CEMRC Web page in July 1999. The FAS Lane presents the results of air sampling in the exhaust shaft at the WIPP which are updated weekly. This represents the most timely data available to the public concerning environmental parameters in the vicinity of the WIPP. A Laboratory Information Management System module was implemented and customized for the management and reporting of data for the FAS Lane.

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

During 1999, the CEMRC hosted 18 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. The CEMRC was involved in a variety of other outreach activities including presentations for local civic and professional groups and an exhibit for a border environmental technology conference (Appendix F). As described in a later section, over 400 volunteers

from the local community have participated in the "Lie Down and Be Counted" project. In addition, CEMRC scientists provided leadership in a variety of professional and scientific organizations and meetings (Appendix G).

In July 1999, the CEMRC distributed the first issue of its newsletter, *The Monitor*. The newsletter summarized progress achieved in the Lie Down and Be Counted project, described the FAS Lane and other aspects of the CEMRC web site, and provided general information about the CEMRC. Over 2000 copies of the newsletter were distributed to local residents and regular recipients of CEMRC reports. Future newsletters will be issued twice annually.

The CEMRC hosted a visiting scientist during 1999 (Dr. Xioaye Zhang, State Key Laboratory of Loess & Quaternary Geology, Shaanxi Province, People's Republic of China). Efforts are still in progress for a fellowship placement by the International Atomic Energy Agency.

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

CEMRC scientists generated 11 proposals, pre-proposals and contract modifications during 1999 (Appendix H). New or expanded funding was achieved on five projects totaling over \$600,000, three proposals are pending, and four proposals were not funded. A total of 11 projects (external to the CEMRC) were in progress during 1999, with a combined value of approximately \$1.6 million. These projects represent a wide array of activities, and they have resulted in significant expansion and diversification of the scientific program. During 1996-1999, CEMRC has received funding from a total of 14 different federal and private sponsors.

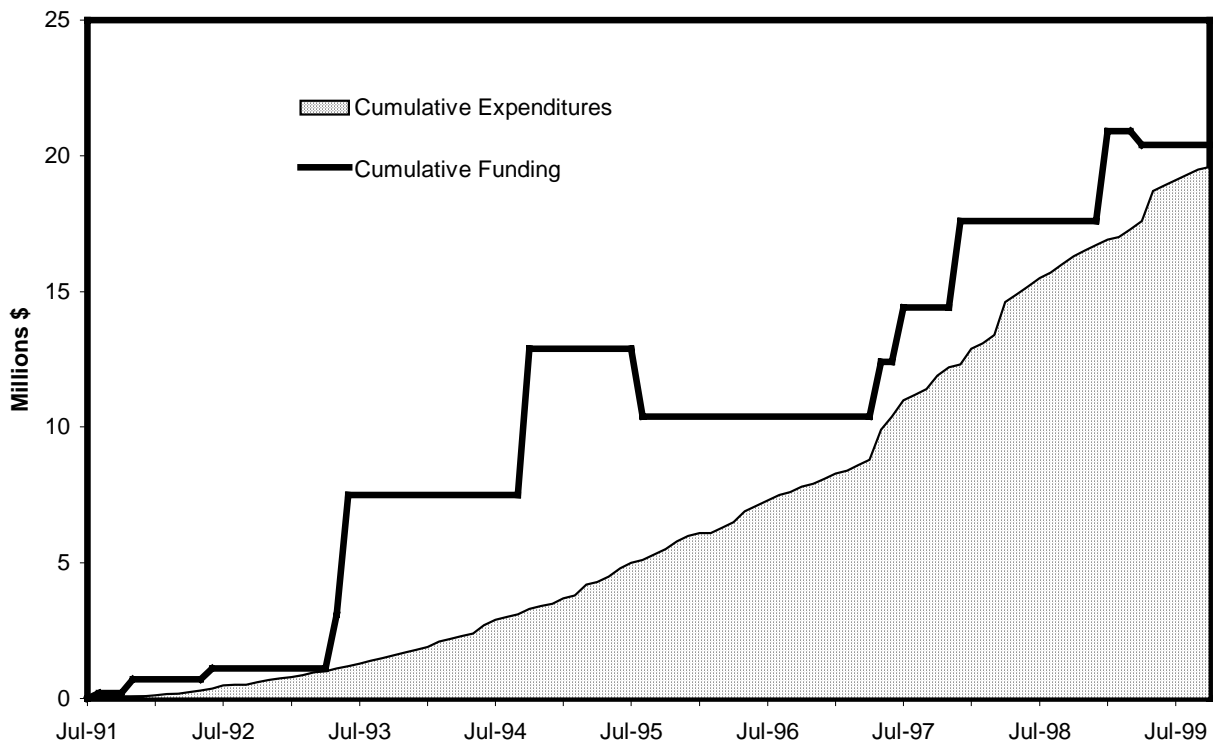
## **9. Implement programs to offer technical training in specialized research techniques and methodologies and to involve CEMRC resources and personnel in providing educational opportunities for students nationwide.**

During 1999, a total of six undergraduate students worked in laboratory aide and technician positions at the CEMRC; these positions



provide training and basic skills development relevant to the position assignments. Two CEMRC scientists hold Graduate Faculty appointments at NMSU, and CEMRC provided analytical support for a graduate research project in the NMSU Department of Fishery

and Wildlife Sciences. CEMRC staff presented lectures on chemical risk assessment for two short courses for environmental professionals (Appendix D). Nine major presentations and special programs were provided for student groups (Appendix F).



**Figure 1. History of CEMRP Funding and Expenditures**

**Table 1. Listing of CEMRC Staff as of 31 December 1999**

<b>Name</b>	<b>Position</b>
Arimoto, Richard	Senior Scientist-Environmental Chemistry
Brown, Becky	Fiscal Specialist II
Castillo, Rick	Technician III-Environmental Chemistry
Chatfield, Randy	Programmer/Analyst I
Conley, Marsha	Director
Cooper, Andy	Technician II-Field Programs
Curbello, Aaron	Technician III-Field Programs
Kirchner, Thomas	Senior Scientist-Informatics & Modeling
Litinsky, Lidia	Quality Assurance Manager
Lynch, Sherry	Technician IV-Science
Madison, Tom	Project Manager
Maung, Okka	Assistant Scientist-Radiochemistry
Monk, James	Technician IV-Internal Dosimetry
Nesbit, Curtis	Associate Health Physicist
Sage, Sondra	Assistant Scientist-Field Operations
Schloesslin, Carl	Assistant Scientist-Radiochemistry
Schloesslin, Cheryl	Assistant Scientist-Environmental Chemistry
Schoep, David	Science Specialist-Internal Dosimetry
Staley, Jeremy	Computer Technician II-Informatics & Modeling
Stevens, Thaddeus	Programmer/Analyst I
Stewart, Barry	Associate Scientist-Radiochemistry
Stroble, Carolyn	Buyer Specialist I
Webb, Joel	Manager, Program Development
Webb, Scott	Associate Scientist-Radiochemistry
Yahr, Jim	Assistant Scientist-Field Operations
York, Larry	Technician II-Radiochemistry
Young, Karen	Administrative Secretary II

## WIPP Environmental Monitoring Project

### Project Concept

As defined in the original grant language, 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.

A detailed description of the WIPP EM concepts, sampling design and baseline studies is presented on the CEMRC web page. The following is a brief summary of 1998-1999 activities for each major environmental medium in the WIPP EM. Because mixed waste has not yet been deposited in the WIPP, current analyses of non-radiological constituents are a continuation of baseline sampling.

**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 the presence of radiological contaminants in the region of the WIPP that differ from observed baseline levels for the targeted analytes.**

### Aerosols

Aerosol sampling is conducted at four 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) and a site approximately 19 km southeast (upwind) of the WIPP (Cactus Flats station) (Fig. 2).

Continuous sampling of aerosols was conducted throughout 1999 using the same instruments, frequencies and locations as were previously established in the baseline phase.

Analyses of aerosol samples collected through July 1999 for non-radiological constituents were completed and reported herein. Analyses of radioactive constituents in aerosol samples from the On-Site, Near Field and Cactus Flats stations collected during 1997-1999 are in progress, and the results will be posted on the CEMRC web site in May 2000. Web site posting of results of radiological and non-radiological analyses of aerosol samples collected in the WIPP exhaust shaft (FAS) began in July 1999, and are updated weekly. A summary of these data is presented herein.

For the long-term monitoring phase of the project (2000 and subsequent years), essentially the same aerosol sampling design will be maintained, except that sampling used for non-radiological constituents will be reduced to only one type of sampler (either low-volume or dichotomous samplers).

### Soils

Soil sampling is conducted within a 166 km<sup>2</sup> area centered on the WIPP operations facility, and at a comparable area encompassing the Cactus Flats aerosol sampling station. Within each of these two areas, samples are collected at 16 locations positioned in concentric rectangular grids (Fig 2). For baseline studies at each of the 16 locations in each area, samples were collected at three randomly selected sites within 50 m of the location's reference point. The resulting data represented 96 discrete samples that provided estimates of variability at the small-scale (between samples within a 0.0025 km<sup>2</sup> area), medium-scale (among locations within each 166 km<sup>2</sup> area) and large-scale (between the two sampling areas located approximately 19 km apart). In 1998, the full complement of 96 soil samples was collected during March-April. A complete characterization of radiological and non-radiological constituents was completed for the 1998 samples and is reported herein.

In 1999, another full suite of 96 samples was collected just prior to the first receipt of waste at the WIPP. These samples have been archived for future reference materials. For the long-term monitoring phase of the project (2000 and subsequent years), one soil sample

will be collected at each of the 32 locations once annually, during January-February. The limitation of soil sampling to one period annually is based on the assumption that any input of contaminants to surface soils from WIPP releases would occur via aerosol deposition, and since aerosol sampling is conducted continuously, more frequent soil sampling is not warranted unless there was evidence of a contaminant increase in aerosols.

### **Surface Water and Sediments**

The WIPP EM incorporates studies at three reservoirs on the Pecos River, which is the major perennial fresh water system closest to the WIPP that has extensive human usage. The three reservoirs are (1) Brantley Lake, located approximately 40 miles northwest of the WIPP, (2) Lake Carlsbad, located in Carlsbad and approximately 25 miles northwest of the WIPP and (3) Red Bluff Lake, located approximately 30 miles southwest of the WIPP. A preliminary study of selected radionuclides in sediments at Brantley Lake was conducted in spring 1997, and results were reported in the CEMRC 1997 Report. During 1998, sediment and surface water samples were collected during the winter and spring, and results were reported in the CEMRC 1998 Report. The first monitoring phase samples were collected in the summer of 1999.

Analyses of the 1999 surface water and sediment samples for non-radiological constituents were completed and are reported herein. Analyses of selected alpha-emitting radionuclides in 1998 (baseline) sediment samples and tests for Pu in 1998 surface water samples were also completed and are reported herein. Radiological analyses of 1999 (monitoring phase) surface water samples were completed and are reported herein. Radiological analyses of 1999 sediment samples are scheduled to be completed and reported by August 2000.

Because of the distance between the WIPP site and these reservoirs, the potential risk of direct contamination of the reservoirs by releases from the WIPP is relatively low compared to other media, and sampling in subsequent years will continue to be conducted once annually in the summer.

### **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 ten miles of the WIPP is also sampled.

During initial baseline studies in 1996-1998, the drinking water samples were subjected to a suite of analyses for over 150 analytes, including those that are regulated under the Safe Drinking Water Act, and contaminants known or suspected to be present in the WIPP wastes. Radioanalyses of drinking water conducted during 1997-1998 (previously reported) were unable to detect Pu or Am in any of the samples collected, using traditional alpha spectrometry.

A set of drinking water samples collected during 1998 were submitted for analyses by thermal ionization mass spectrometry (TIMS) (a more sensitive radioanalytical technique) and Pu was not detectable in any of the water samples (reported herein). Based on these results, subsequent radiological analyses have applied standard alpha spectroscopy detection limits achievable with 3-L samples. The first monitoring phase drinking water samples were collected in the summer of 1999, and results of radiological analyses are reported herein. Analyses of non-radiological constituents for the 1999 samples were also completed and are reported herein. The six drinking water supplies will continue to be sampled once annually for selected radiological and inorganic testing.

### **Biota**

Studies of biota for the WIPP EM have focused on native vegetation because the vegetation is consumed by beef cattle, and consumption of beef from cattle pastured in the vicinity of the WIPP could serve as an exposure pathway to humans for contaminants released from the WIPP. During baseline stud-

ies, vegetation samples were collected from a total of six species of plants that serve as preferred forage species for cattle during at least some portion of the year. During 1997-1998 baseline studies, vegetation was sampled twice annually during the two major periods of new growth for native vegetation (March-May and August-October). Six samples of each of three species (contingent on availability) were collected during each sampling period from selected sites on the sampling grid surrounding the WIPP (which encompasses the Near Field aerosol sampling station). Due to absence of new growth resulting from lack of rainfall, no vegetation was collected during spring 1999. During late summer 1999, the first monitoring phase samples were collected, consisting of a single sample of several grass species from each of the 32 soil sampling locations. Radiochemical analyses of the baseline and initial monitoring phase samples will be carried out during 2000, followed by web page posting and inclusion in the CEMRC 2000 Report. Vegetation sampling will continue to be conducted once annually during operational monitoring phases, focusing on species collected during baseline studies.

Additional studies are in progress to evaluate the effectiveness of expanding the biota sampling for radionuclides to include arthropods. Arthropods were collected at the Cactus Flats station during spring through fall in 1998 (baseline) and at Near Field and Cactus Flats stations during spring through fall in 1999. Comparative radioanalyses of the samples are planned during 2000, to evaluate the effectiveness of this environmental component in long-term monitoring.

### **Human Population**

The "Lie Down and Be Counted" 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 many 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

margin of error for comparisons between major population ethnicity and gender categories as identified in the 1990 census. The minimum sample size threshold was achieved for the major categories early in 1998, and continued baseline sampling resulted in radiobioassays completed for 367 individuals before the first receipt of waste at the WIPP, reducing margin of error ranges to a maximum of 5-7% for any category. Radiobioassays of the original volunteer cohort began in July 1999 to achieve a complete cohort recount by July 2001. A minimum of 100 new volunteers will be incorporated each year to establish new study cohorts.

Results of the Lie Down and Be Counted project through 1 October 1999 are reported herein, and are updated quarterly on the CEMRC Web site.

### **Meteorological Monitoring**

Fully automated meteorological stations are operated by the CEMRC at the Near Field aerosol station and the Cactus Flats aerosol station. Details concerning the sensors and operation of the equipment and a summary of the last year's meteorological patterns are presented herein.

### **Management of WIPP EM Project**

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 1999 key performance indicators is summarized in Appendix I. Nine indicators were completed on time and three indicators were delayed but completed prior to year-end. Five

out of 17 indicators were not completed, with 0-90% progress on each. Key performance indicators for 2000 have been identified to

serve as the basis for the 2000 WIPP EM project schedule (Table 2).

**Table 2. Key Performance Indicators for 2000**

Focus Area	Key Performance Indicator
Aerosols	1. Continue concurrent high-volume and low-volume/dichotomous sampling at current three locations through 2000
	2. Continue collection of daily FAS samples in WIPP exhaust shaft through 2000
Soils	3. Collect samples at current 32 locations during January-February 2000
Meteorology	4. Continue concurrent operation of sampling stations at two current sites through 2000
Drinking water	5. Collect samples from six sources during March-April 2000
Sediment and surface water	6. Collect samples from three reservoirs during June-July 2000
Biota	7. Collect vegetation samples from at least six locations during fall 2000
Human studies	8. Complete repeat counts for approximately ½ of original volunteer cohort, and initial counts for a minimum of 100 new volunteers
Radioanalyses	9. Complete analyses of all pre-2000 aerosol, sediment, surface water, drinking water and vegetation samples by October 2000
	10. Complete analyses of soil, aerosol, sediment, surface water and drinking water samples (collected through June 2000) by October 2000
	11. Continue FAS sample analyses to meet weekly and quarterly posting schedule
Non-radiological analyses	12. Complete analyses of representative subset of 2000 low-volume aerosol, soil, sediment, surface water and drinking water samples within three months after each sample collection
	13. Continue FAS sample analyses to meet weekly and quarterly posting schedule
Data management and dissemination	14. Post results of radioanalyses of pre-2000 samples within two months after completion of analyses of each set of samples
	15. Post results of non-radiological analyses of 2000 samples within two months after completion of analyses of each set of samples
	16. Issue CEMRC 1999 Report; post report and background data to CEMRC web site by March 2000
	17. Issue newsletters in March and September, 2000
	18. Submit manuscript for publication by February 2000 on baseline characteristics of soils
	19. Submit manuscript for publication by July 2000 on baseline characteristics of aerosols

## 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 CEMRC has adopted a general quality assurance policy (Appendix J) that includes development and implementation of appropriate standards, performance assessment, quality improvement, provision of infrastructure, professional staff development, personal accountability and commitment to compliance.

The CEMRC's quality assurance policy and implementation plans recognize that there are distinctions between standard analytical activities and experimental research settings. For experimental research settings, there are frequently few if any recognized analytical standards or procedures for the analyses of interest, and a major task is to develop such procedures, or to modify the application of standard procedures for new media. Likewise, research sampling designs are typically unique to the underlying scientific hypotheses, and therefore may not follow any standardized external formats. Therefore, the quality control measures applied to research contrast with those applied in programs driven by regulatory requirements, where the sampling frequency and methodologies and the analytical procedures typically are spelled out by various compliance guidelines.

In the WIPP Environmental Monitoring project, the CEMRC's strategy is to develop a set of independent data for a variety of parameters of interest, frequently using sampling and analyses that are different from those dictated by the regulatory requirements that govern the WIPP's certification and operation. In many cases, these data will target a larger suite of parameters or lower detection limits than are of concern from a regulatory perspective. Although this approach may include some sampling and analyses similar to those conducted by other groups associated with the WIPP, other activities are unique to the CEMRC's projects.

### **Personnel**

Program managers provide training to laboratory and field workers in methodologies, general laboratory protocol and maintenance routines and good safety practices. CEMRC laboratory and technical support staff receive specialized training for operation of specific equipment or systems, generally offered through equipment vendors. To support continued professional development, staff members are also provided opportunities for membership and participation in professional organizations, including attendance at conferences and workshops. Access to current scientific literature is provided through a current publications bulletin, a variety of journal subscriptions and inter-library loans.

### **Regulatory Compliance**

To promote good health and safety practices in the laboratories, the CEMRC maintains a Chemical Hygiene Plan and associated training of personnel, in compliance with the requirements of 29 CFR 1910.1450, "Occupational Exposure to Chemical Hazardous Chemicals in Laboratories." A Hazard Communication Plan and associated training are also maintained for all employees, in compliance with requirements of 29 CFR 1910.200. A Chemical Hygiene Officer and Hazard Communications Coordinator are responsible for management of the chemical and laboratory safety programs, including maintenance of chemical inventories, periodic audits and management of any hazardous wastes generated by laboratory activities.

The CEMRC is a conditionally-exempt small quantity generator of hazardous wastes, as defined and regulated under the Resource Conservation and Recovery Act. Hazardous waste thus generated is disposed of through licensed treatment, storage and disposal facilities. Based on current chemical inventories, the CEMRC is exempt from the reporting requirements in Section 313 of the Emergency Planning and Community Right-to-Know Act. The CEMRC has had no spills of hazardous substances that exceeded the reportable quan-



tity limits under the Comprehensive Environmental Response, Compensation and Liability Act. The CEMRC currently has no air contaminant emissions subject to regulation under the Clean Air Act, and no wastewater discharges subject to regulation under the Clean Water Act beyond normal sanitary sewer discharges.

Use of radioactive materials is governed by the CEMRC's Radioactive Materials License, issued by the New Mexico Environment Department. A Radiation Control Manual and Implementation Plan and associated training are provided for staff who deal with radioactive materials. A Radiation Safety Officer is responsible for management of the radiation safety program, including maintenance of a radioactive materials inventory, periodic radiation contamination surveys, radiation safety audits and management of any radioactive waste generated by laboratory activities. The CEMRC generates a small amount (<100 lb) of solid, low-level radioactive waste annually, which is disposed of through a licensed commercial disposal facility.

#### **Field Sampling Program Quality Assurance**

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 preliminary plans were developed and documented. After the activity was completed, the plan was 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 were operated to meet an EPA standard of  $1.13 \text{ m}^3 \text{ min}^{-1}$ , 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. Original logbooks and field data forms are kept on file in the program manager's office. Most data collected in written form are also entered in electronic files, and electronic copies are cross-checked 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 adapt to special project needs. Calibrations are either carried out by equipment vendors or by CEMRC personnel using certified calibration standards. Records of calibration and maintenance are maintained in instrument-specific files in the program manager's office.

#### **Environmental Chemistry Program Quality Assurance**

The analytical methods employed in the environmental chemistry program at CEMRC are based, when applicable, on various standard procedures (EPA, 1983, *Methods for Chemical Analysis of Water and Wastes*, EPA/600/4-79-020; EPA, 1997, *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*; EPA/SW-846; American Public Health Association, 1981, *Standard Methods for the Examination of Water and Wastewater*, 20<sup>th</sup> Edition).

For the WIPP EM, an ion chromatograph (IC) was used to determine the concentrations of a suite of major ions in water samples and aqueous extracts of all media sampled except vegetation (Table 3). The atomic absorption spectrometer (AAS) and inductively coupled plasma-mass spectrometer (ICP-MS) were used to analyze aqueous or acid extracts of samples (excluding vegetation), depending on the particular question or issue being addressed.

For some matrix/analyte combinations, appropriate external standard procedures do not exist, and CEMRC has developed specialized standard procedures to meet the needs of the WIPP EM. A set of standard operating procedures and a formal quality assurance plan have been developed and implemented for the inorganic analyses performed at CEMRC. A summary of the quality assurance/quality control procedures applied by the environmental chemistry program for WIPP EM studies is presented in Appendix K. During November 1999, the environmental chemistry program began analyses for performance testing under the National Voluntary Laboratory Accreditation Program, but test results had not been received by the end of the year.

### **Radiochemistry Program Quality Assurance**

During 1999, the CEMRC radioanalytical program participated in two rounds of the NIST Radiochemistry Intercomparison Program (NRIP) and achieved traceability for all analytes reported. The radioanalytical program also participated in the DOE Environmental Measurement Laboratory Quality Assurance Program (EML QAP), resulting in "acceptable" ratings for 45 results from glass fiber filters, soil, vegetation and water samples.

CEMRC has undertaken an extensive method development and validation project that began in May 1998 and will extend through 2000. New standard procedures have been developed and adopted for WIPP EM analyses for soil samples, water samples and aerosol filters. Method development will continue for sediment and biota during 2000. The completion of this developmental phase will include adoption of a formal quality assurance plan and implementing procedures for radioanalyses in the WIPP EM project. A summary of general quality assurance/quality control procedures used by the radioanalytical program is presented in Appendix L.

### **In Vivo Radiobioassay Quality Assurance**

*In vivo* radiobioassays are performed in accordance with a formal quality assurance plan and related documentation that were devel-

oped to meet the requirements of the Department of Energy Laboratory Accreditation Program (DOELAP) for Radiobioassay. During 1999, CEMRC received performance-testing results from DOELAP for  $^{241}\text{Am}$  and  $^{235}\text{U}$  in lungs and  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in whole body. CEMRC passed all performance criteria with relative biases for  $^{241}\text{Am}$ ,  $^{235}\text{U}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  of -0.14, 3.77, 2.00 and 6.25%, respectively. The DOELAP onsite assessment of CEMRC was performed during the summer of 1999. Through this assessment, CEMRC's radiobioassay quality assurance, quality control and operational practices are scrutinized by national experts in radiobioassay. No deficiencies were determined in the CEMRC program and final accreditation was awarded in late 1999.

During 1999, the CEMRC *in vivo* radiobioassay program participated in the Intercomparison Studies *In Vivo* Program administered by Oak Ridge National Laboratory (ORNL). This program provides quarterly testing for  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{57}\text{Co}$ ,  $^{88}\text{Y}$  and  $^{133}\text{Ba}$  deposited in whole body. From the 1998 annual report issued in the Spring of 1999 (four quarters in total), CEMRC reported values that were within -0.62 to 3.25% of the ORNL known value for all radionuclides.

### **External Laboratory Services**

Some analyses presented herein were carried out by other laboratories through subcontract or fee service arrangements. These include analyses of radiological constituents in sediments, surface water and drinking water samples and analyses of soil texture.

The Soil, Water, and Air Testing Laboratory (SWAT) at NMSU provided analyses of soil texture. The SWAT quality assurance/quality control program is documented in a Quality Assurance Program Plan (QA-QAPP-1).

Duke Engineering & Services (DES) (Bolton, Massachusetts) conducted Radioanalyses of alpha-emitting radionuclides in sediments reported herein. DES maintains a quality assurance program as documented in the DES Environmental Laboratory Quality Assurance Plan (Manual 100). DES has achieved acceptable performance for analyses of environmental samples in the DOE EML

QAP, and maintains traceability to NIST through the NEI/NIST Measurement Assurance Program.

Los Alamos National Laboratory, Chemical Science and Technology Division conducted analyses of  $^{239}\text{Pu}$  in surface water and drinking water samples. These analyses employed distillation, separation, purification and electrodeposition, followed by thermal ioniza-

tion mass spectrometry, as described by Perrin et. al., (1985, *Int. J. Mass Spectrometry and Ion Physics*, 64, 17). Use of this technique was recently successful in documenting long-distance migration of Pu in ground water at the Nevada Test Site (Kersting, A.J., et. al. 1999, *Nature*, 397, 56).

**Table 3. Inorganic Analyses Conducted at CEMRC**

Type of Sample	IC (Anions)	IC (Cations)	Hydrides (As, Sb and Se) and Hg, Flow Injection AA	Trace Elements (for ICP-MS & AA)
Air	Filter extraction with DI water and isopropanol	Filter extraction with DI water and isopropanol		Aqueous extract Total dissolution
Drinking and Natural Waters	Syringe filtration with direct injection		Syringe filtration with direct injection	Dissolved Total recoverable
Soils	Aqueous extract		Total recoverable	Total recoverable
Sediments	Aqueous extract		Total recoverable	Total recoverable



**WIPP ENVIRONMENTAL MONITORING DATA SUMMARIES**

## Meteorological Conditions in the Vicinity of the WIPP Site

### Methods

CEMRC operates two identical meteorological towers at sampling sites in the vicinity of the WIPP (Fig. 2). The Near Field site is located approximately 1 km northwest of the WIPP site at an elevation of 1088 m (latitude 32°22'40.385"N; longitude 103°47'55.425"W). The Cactus Flats site is located approximately 19 km southeast of the WIPP site at an elevation of 1041 m (latitude 32°13'05.451"N; longitude 103°41'42.583"W).

Each station consists of a 10-meter tower equipped with sensors for temperature, relative humidity, barometric pressure, solar radiation, Ultra-Violet B (UVB) radiation, wind speed and direction and vertical wind speed. Data are collected every second, with averaging times of ten minutes. In addition, the maximum wind speed and total precipitation occurring over the ten-minute averaging period are recorded.

Temperature, relative humidity and all wind parameters are measured at a height of 10 m above the surface. Precipitation, barometric pressure solar radiation and UVB are measured at heights of 0.4, 1, 2 and 3 m, respectively. The barometric pressure sensors are adjusted for temperature, but are not referenced to mean sea level. The solar radiation sensors (pyranometers) measure the energy flux per unit area ( $\text{Wm}^{-2}$ ) of both direct and diffuse sky radiation. The UVB sensor measures direct and diffuse UVB in the 280-320 nm band.

The data are stored in electronic dataloggers and downloaded twice weekly. Once downloaded, the data are screened for outliers and other anomalies and uploaded to a main database. Performance checks of the sensors are conducted quarterly, and sensors are recalibrated at least annually.

This report summarizes meteorological data collected over the 12 month period from December 1998 through November 1999. In addition, data collected at the sites from 1 December 1997 through 30 November 1998

(12 month period) are compared with data from the same time interval during 1998-1999.

### Results

For the 1999 sampling period, data recovery exceeded 97% for all sensors, except the UVB sensor at Cactus Flats. Data recovery was slightly lower at Cactus Flats during the month of June when a nearby lightning strike disabled several of the sensors. In addition, the Cactus Flats UVB sensor failed in April and was undergoing repairs and recalibration during April through July. Other short-term (typically less than two hours) data losses occurred throughout the year due to sensor maintenance, repair, performance testing and malfunction.

Averaged over the year, winds were from a southeasterly direction (E, ESE, SE and SSE quadrants, inclusive) 49% and 48% of the time at the Cactus Flats and Near Field sites, respectively (Fig. 3). However, there were some distinctive seasonal variations in wind direction (Figs. 4-5). Wind direction was highly variable during the winter and spring (December through May) when compared with the summer and fall (June through November). During summer, wind from the southeastern quadrant occurred over 50% of the time, but dropped to less than 40% during the winter. The inter-annual and intra-annual variability in wind direction are important parameters in modeling dispersion pathways for potential airborne releases from the WIPP.

Wind velocities were very similar between sites. Wind velocities (10-minute means) were less than  $5.4 \text{ m s}^{-1}$  over 72% of the time, with speeds frequently from  $3.1$  to  $5.4 \text{ m s}^{-1}$ . Calm periods (wind velocities  $< 0.1 \text{ m s}^{-1}$ ) occurred less than 1% of the time over the year. Wind velocities  $> 5.4 \text{ m s}^{-1}$  occurred less than 28% of the time, but were more frequent during the spring, and typically came from west and west-northwest. The highest wind velocities recorded at each site were  $27.1 \text{ m s}^{-1}$  (62 mph) on 5 September, and  $28.0 \text{ m s}^{-1}$  (64 mph) on 24 May, at the Near Field and Cactus Flats sites, respectively.

Air temperatures at Near Field ranged from  $-10.9$  to  $41.5$  °C and from  $-10.9$  to  $41.0$  °C at Cactus Flats. The maximum temperatures were recorded on 30 June at Cactus Flats, and 1 July at Near Field. The lowest temperatures were recorded on 26 December 1998 at both locations. The annual mean temperatures were  $18.5$  °C and  $18.0$  °C at Near Field and Cactus Flats, respectively. At both locations, December was the coldest month (mean =  $7.5$  °C at Near Field; mean =  $7.2$  °C at Cactus Flats) and August was the hottest month (mean =  $28.4$  °C at Near Field; mean =  $28.1$  °C at Cactus Flats) (Fig. 6).

The annual mean relative humidity at Near Field was 43% and ranged from 4 to 101%. Humidity at the Cactus Flats site was very similar to Near Field, averaging 44% and ranging from 4 to 101%. Mean relative humidities were lowest when temperatures peaked in late spring and early summer (Fig. 7). It should be noted that the accuracy of the relative humidity sensors declines at relative humidities below 12% and above 94%, and readings outside these ranges should be interpreted with caution.

Barometric pressure did not exhibit an obvious seasonal trend at either site (Fig. 8). The annual mean was 893.6 mb at Cactus Flats and 898.3 mb at the Near Field site. The apparent 4.7 mb difference between the sites can be attributed to a 41 m difference in elevation, and this difference is not significant if corrected using standard barometric conversions that incorporate elevation (U.S. Department of Commerce Weather Bureau, 1963, *Manual of Barometry, Vol. 1*, Washington D.C.).

Solar radiation flux ( $\text{Wm}^{-2}$ ) was integrated over daily intervals to calculate total energy received per unit area ( $\text{MJm}^{-2}$ ). As is typical, solar radiation received at the sites peaked in the summer and was lowest during the winter months (Fig. 9). This pattern is due to a combination of increasing solar radiation intensity, less cloud cover and additional hours of daylight during the summer months. Over the year, the daily total solar radiation ranged from 2.5 to 33  $\text{MJ m}^{-2}$  at Near Field and 3 to 32  $\text{MJ m}^{-2}$  at Cactus Flats.

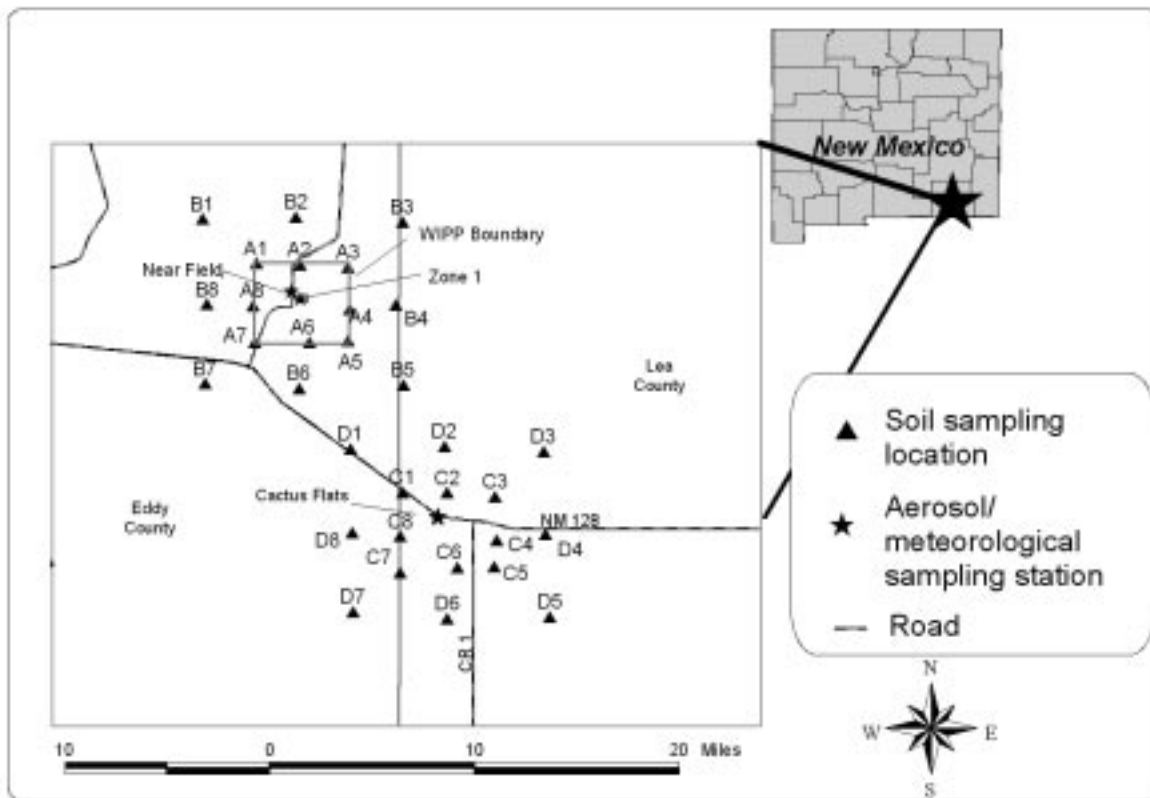
Solar UVB flux ( $\text{Wm}^{-2}$ ) was integrated over daily intervals to calculate total energy

received per unit area ( $\text{MJ m}^{-2}$ ). The UVB radiation followed a pattern similar to that of the total solar radiation (Fig 10).

Over the year, a total of 19.66 cm of precipitation was measured on 43 days at Cactus Flats and 22.88 cm of precipitation was measured on 54 days at Near Field (Fig. 11). At both sites, the month of July had the highest number of days (9 at Near Field, 7 at Cactus Flats) on which precipitation was recorded. At Cactus Flats, December was the month with the highest total precipitation (3.96 cm). In contrast, at the Near Field site, July was the month with the highest total amount of precipitation (5.54 cm).

Overall, 1999 was a slightly wetter year than 1998. In 1998, totals of 17.28 and 12.88 cm of precipitation were recorded at the Near Field and Cactus Flats sites, respectively, between 1 December 1997 and 30 November 1998. Over the same time period in 1999, 22.88 cm of precipitation was recorded at Near Field and 19.66 cm was recorded at the Cactus Flats site. In addition to being wetter, mean annual temperatures were approximately two degrees lower in 1999 than in 1998. Although February, March and April were warmer in 1999, this was offset by approximately 0.3 degrees cooler temperatures in May, June and July of 1999. Given the differences in precipitation and temperature it is not surprising that total solar radiation was also lower in 1999, with Cactus Flats receiving 6449  $\text{MJ m}^{-2}$  in 1999 compared with 7984  $\text{MJ m}^{-2}$  in 1998, and the Near Field site receiving 6575  $\text{MJ m}^{-2}$  in 1999 compared to 7222  $\text{MJ m}^{-2}$  in 1998. In contrast, annual mean wind patterns (velocity and direction) were very similar between years, although some differences appeared in seasonal wind patterns.

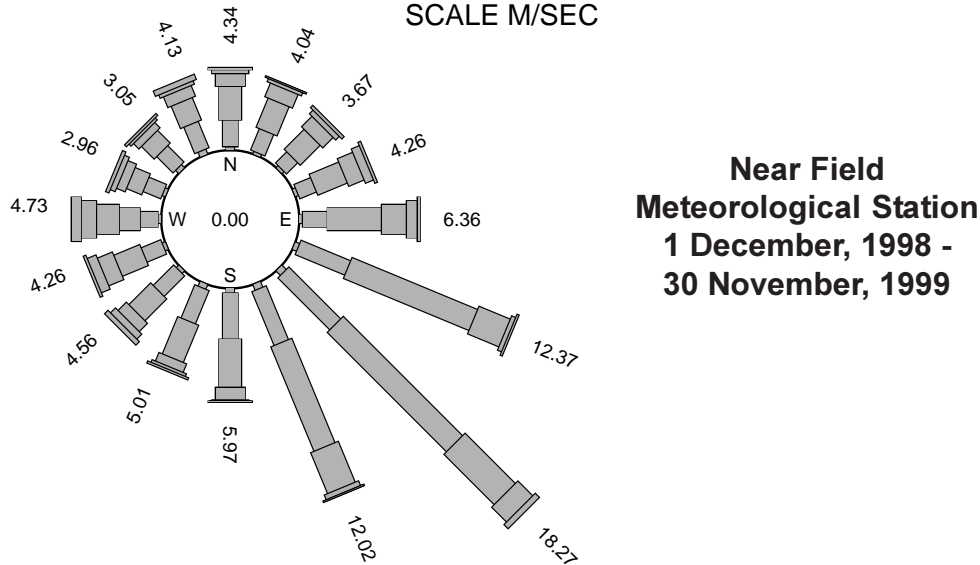
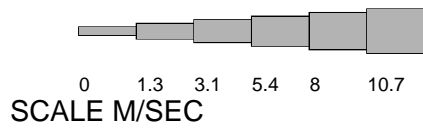
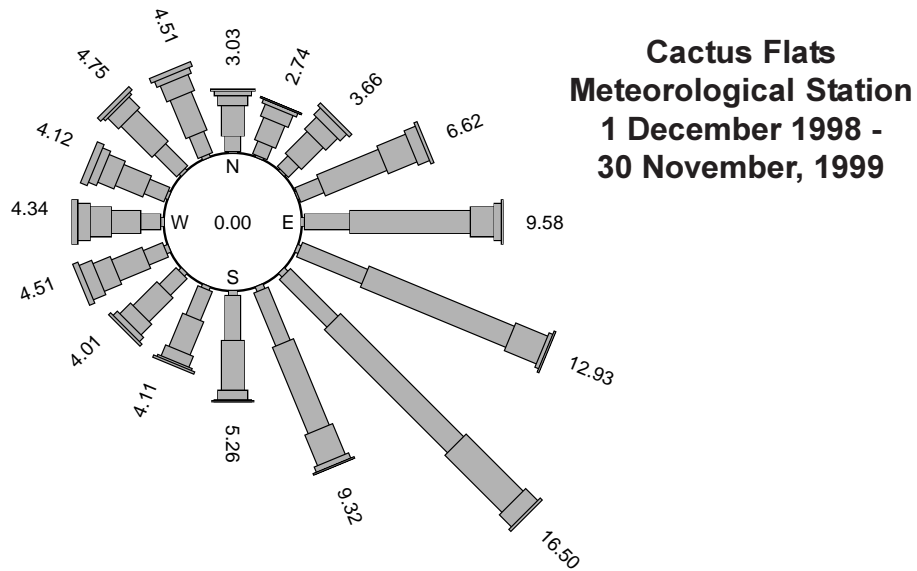
Tables presenting meteorological data summarized herein are available on the CEMRC web site at <http://www.cemrc.org>.



**Figure 2. Sampling Locations in the Vicinity of the WIPP**

Aerosol and meteorological sampling is conducted at Near Field and Cactus Flats.

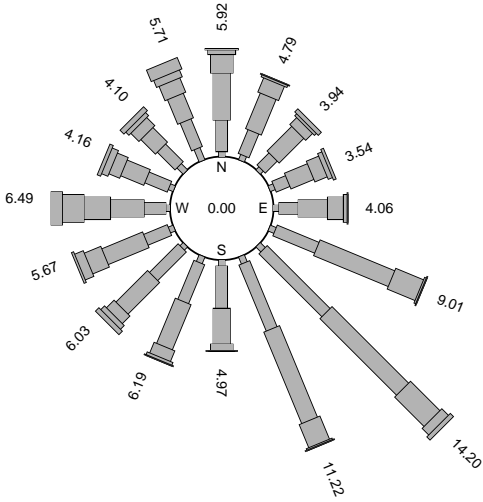




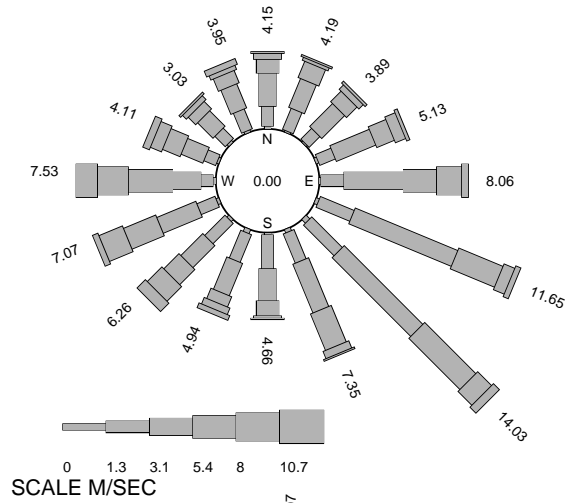
**Figure 3. Annual Wind Roses for Near Field and Cactus Flats**

Value in center of a rose is % time with no recordable wind. Value at outer end of each tube is % time wind blew from the direction of the tube outer opening. Within each tube, segment lengths indicate relative frequency of wind speeds (m sec<sup>-1</sup>) given on scale.

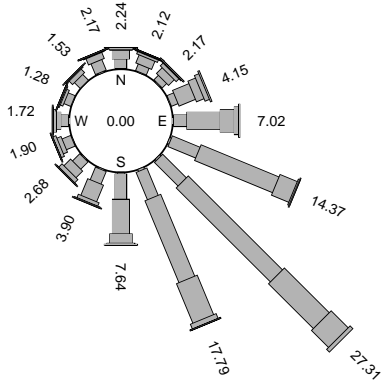
**Near Field  
Meteorological Station  
1 December 1998 -  
28 February 1999**



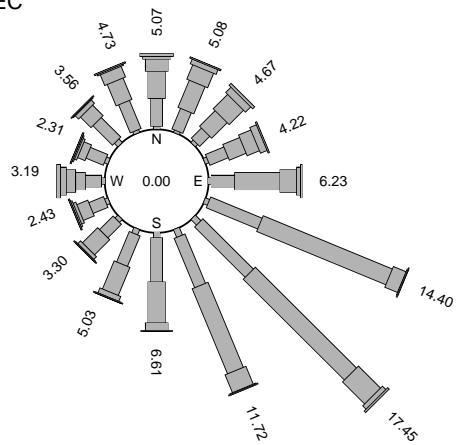
**Near Field  
Meteorological Station  
1 March 1999 -  
31 May 1999**



**Near Field  
Meteorological Station  
1 June 1999 -  
31 August 1999**

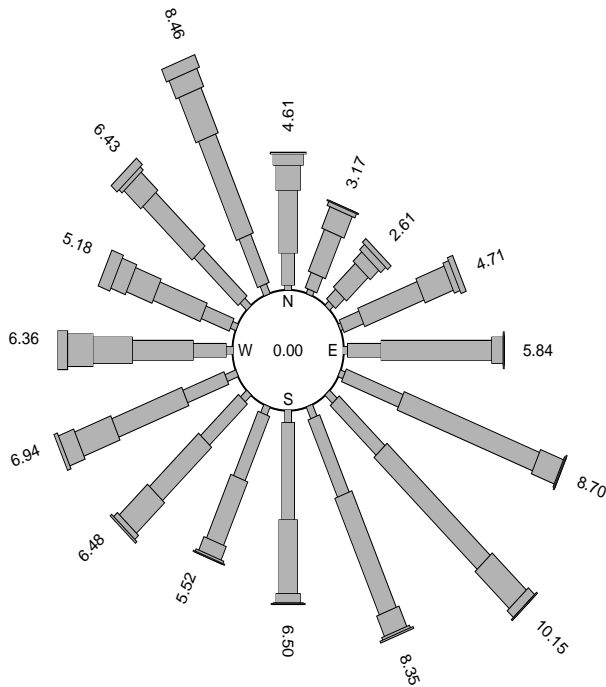


**Near Field  
Meteorological Station  
1 September 1999 -  
30 November 1999**

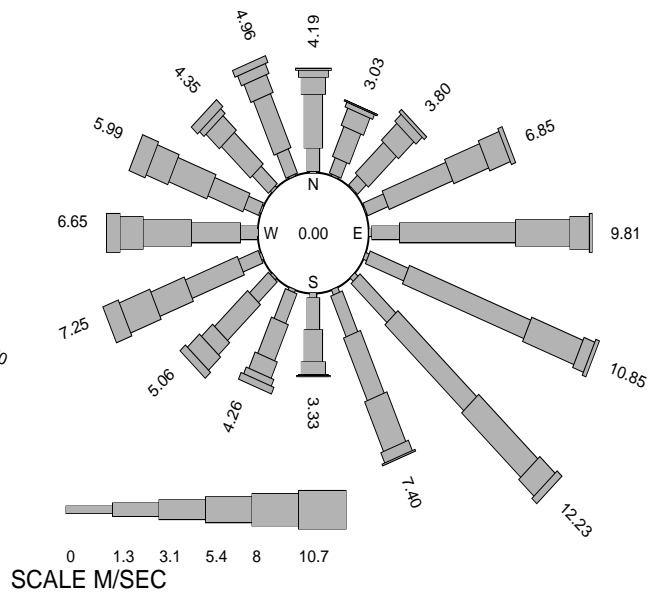


**Figure 4. Seasonal Wind Roses, Near Field**  
See page 21 for explanation.

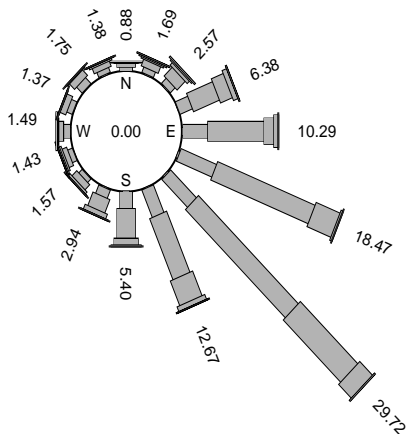
**Cactus Flats  
 Meteorological Station  
 1 December 1998 -  
 28 February 1999**



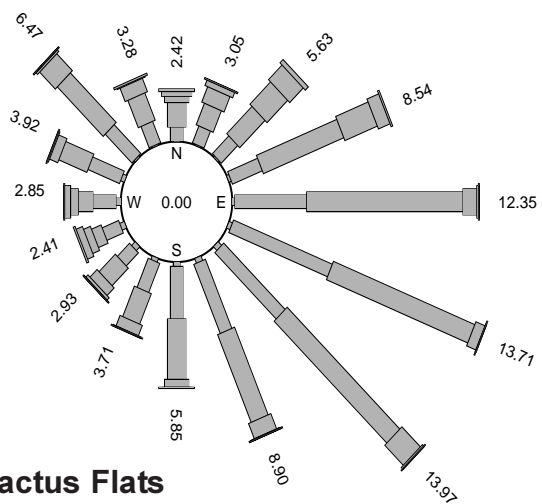
**Cactus Flats  
 Meteorological Station  
 1 March 1999 -  
 31 May 1999**



**Cactus Flats  
 Meteorological Station  
 1 June 1999 -  
 31 August 1999**

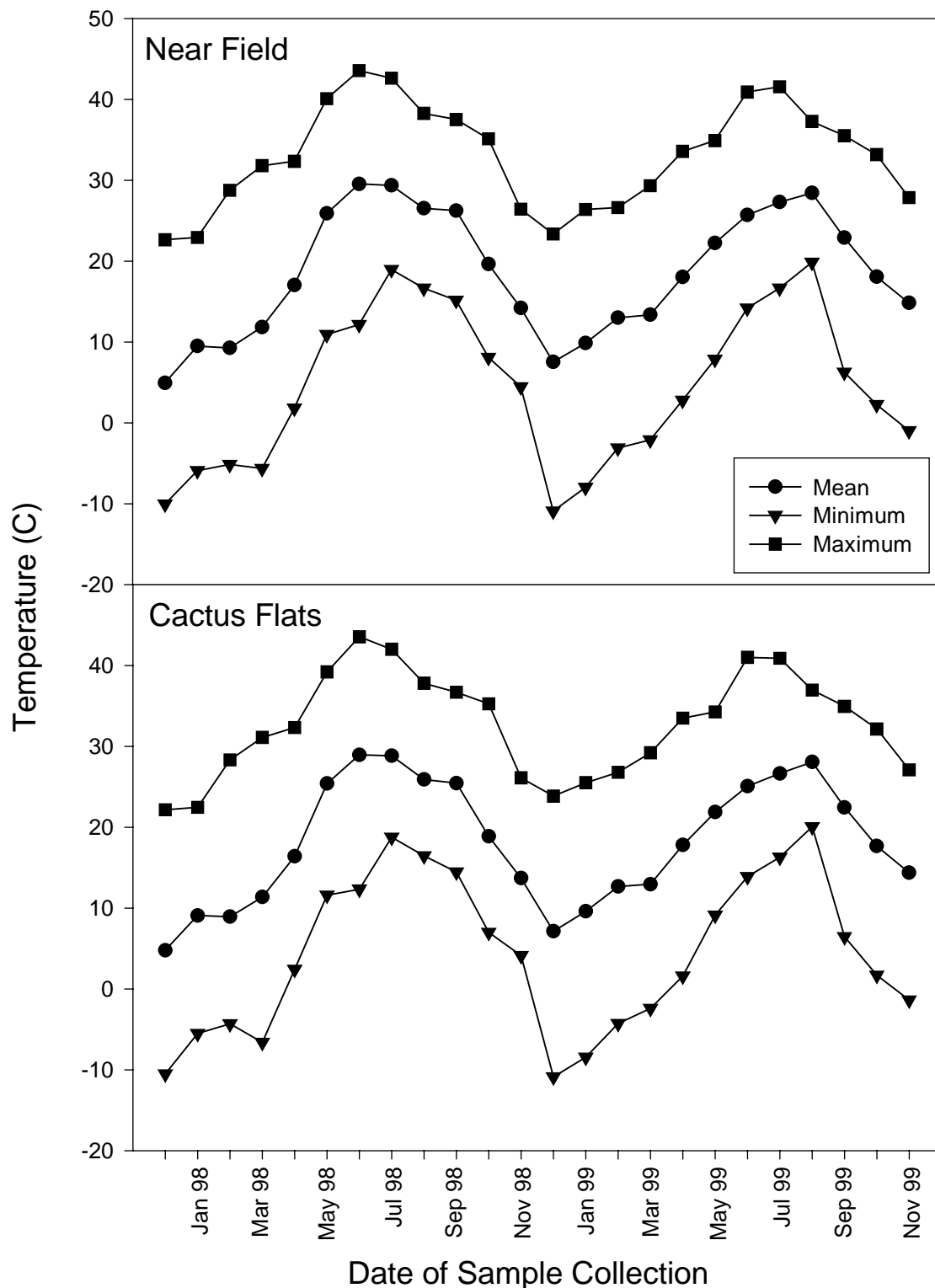


**Cactus Flats  
 Meteorological Station  
 1 September 1999 -  
 30 November 1999**

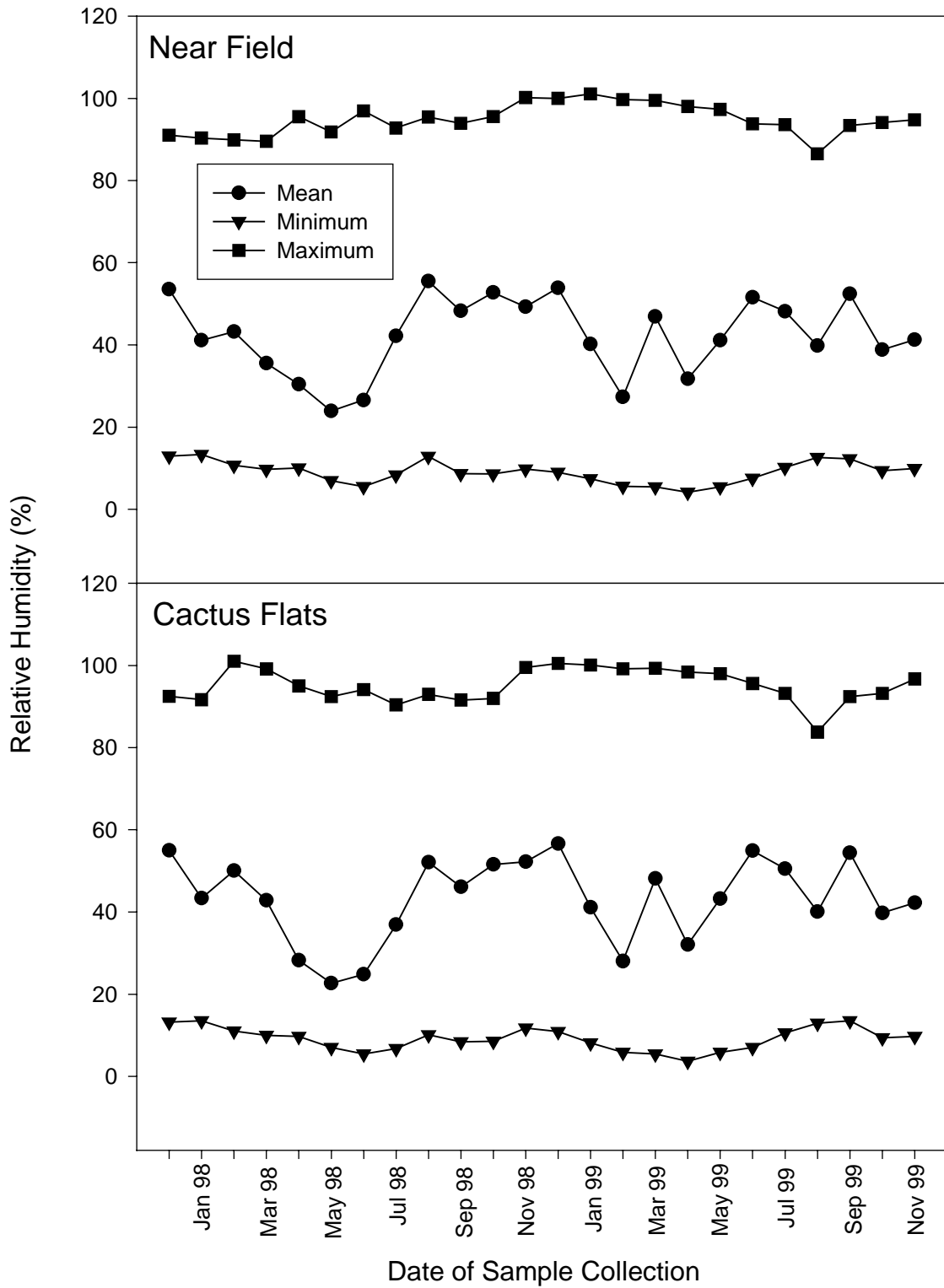


**Figure 5. Seasonal Wind Roses, Cactus Flats**

See page 21 for explanation.

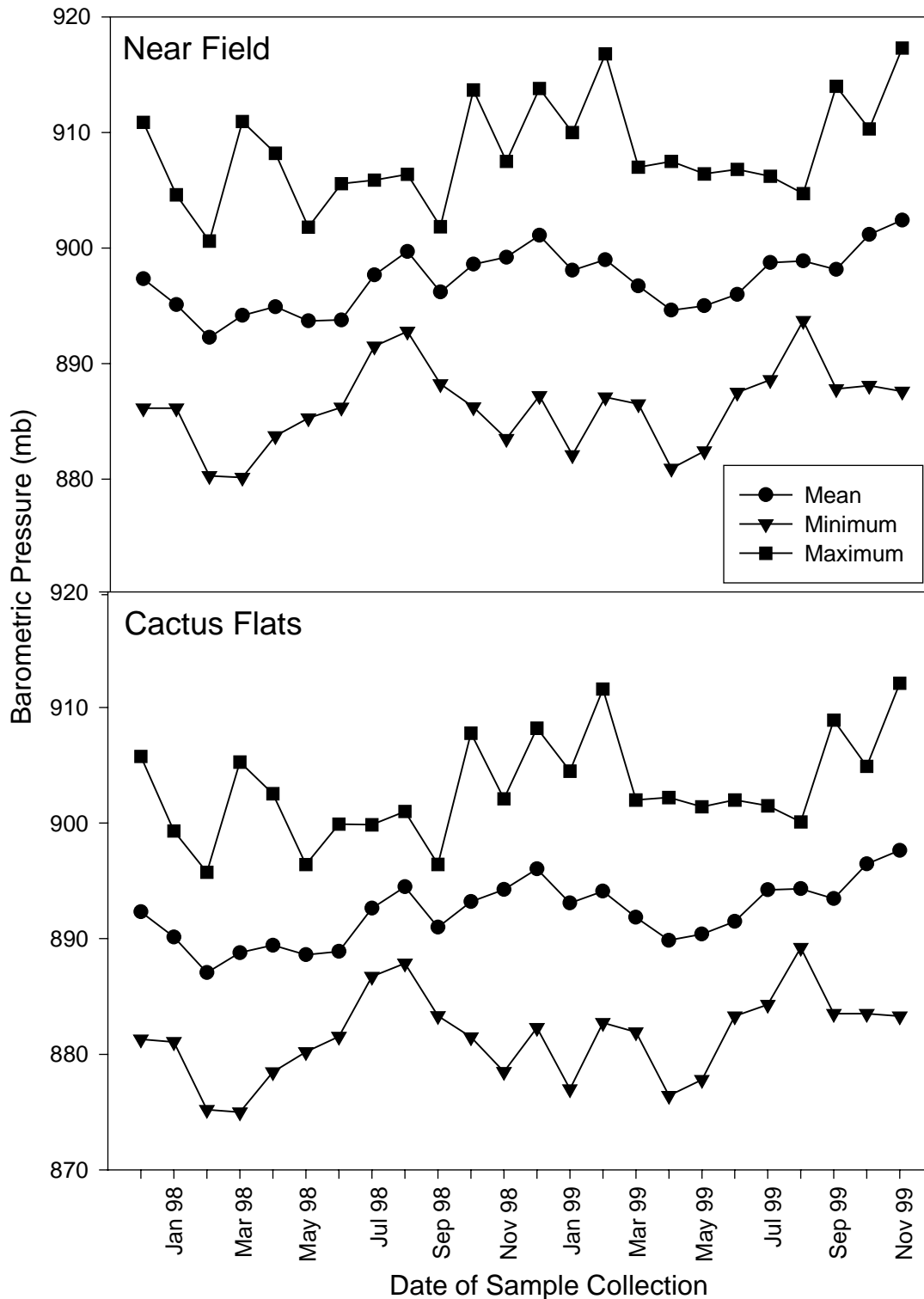


**Figure 6. Monthly Mean, Minimum and Maximum Temperature at Near Field and Cactus Flats during December 1997 - November 1999**

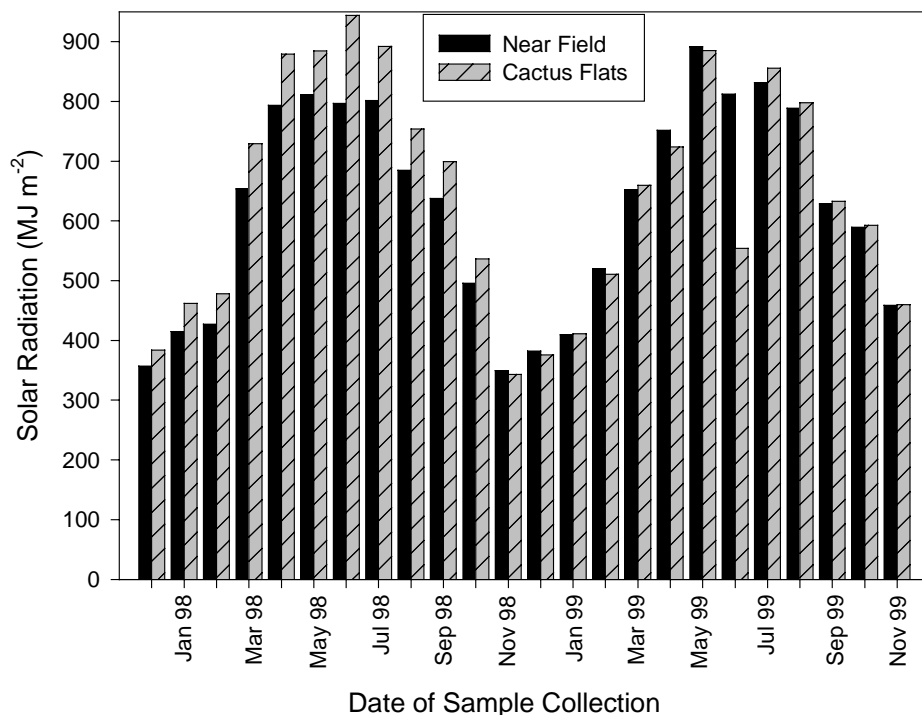


**Figure 7. Monthly Mean, Minimum and Maximum Relative Humidity at Near Field and Cactus Flats during December 1997 - November 1999**

Relative humidity sensor may have reduced accuracy at <12% and >94%.

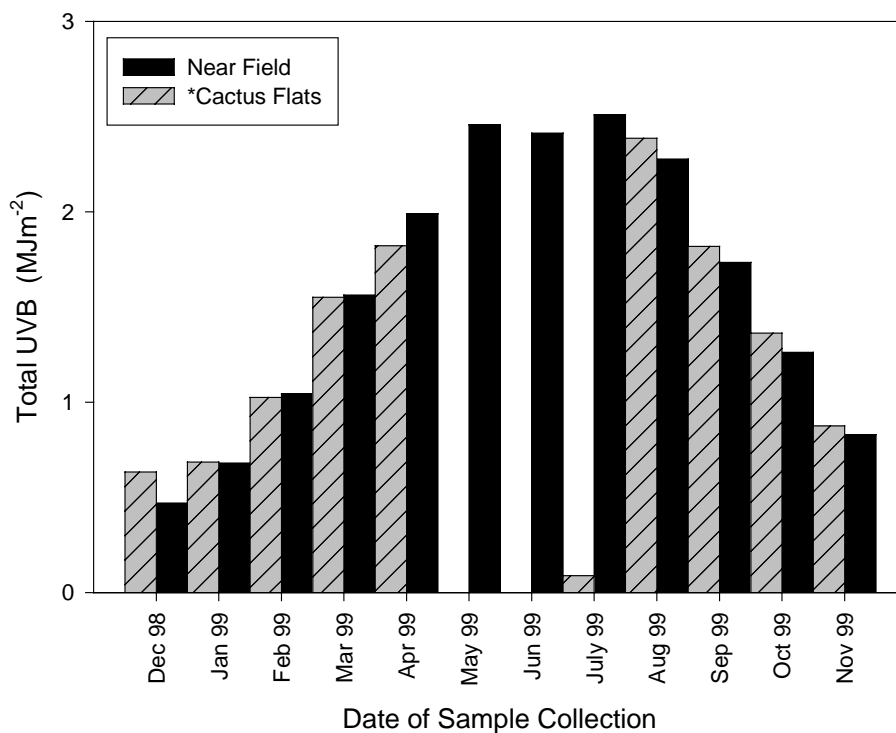


**Figure 8. Monthly Mean, Minimum and Maximum Barometric Pressure at Near Field and Cactus Flats during December 1997 - November 1999**



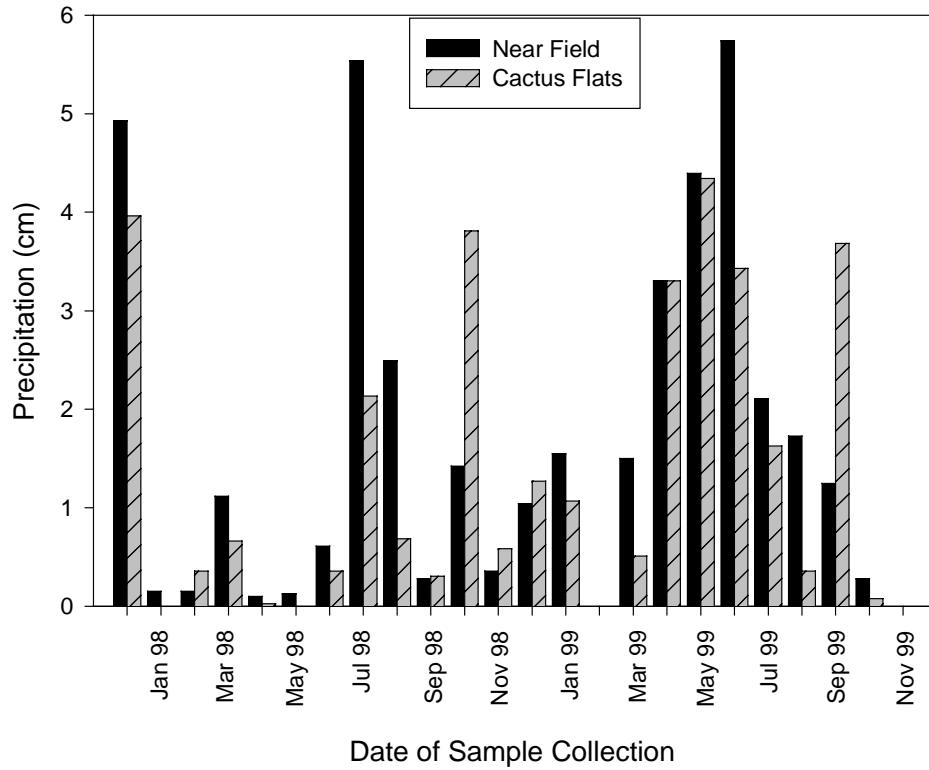
**Figure 9. Monthly Total Solar Radiation at Near Field and Cactus Flats during December 1997 - November 1999**

The low value for June 1999 at Cactus Flats resulted when the sensor was damaged by a lightning strike.



**Figure 10. Monthly Total UVB Radiation Measured at Near Field and Cactus Flats during December 1998 - November 1999**

\*Cactus Flats sensor inoperative during 28 April – 30 July 1999.



**Figure 11. Monthly Total Precipitation at Near Field and Cactus Flats during December 1997 - November 1999**



## Particulate Concentrations and Inorganics in Near-Surface Air

### Introduction

The CEMRC aerosol studies, which are a key component of the WIPP EM project, focus on a transport pathway that could cause the rapid dispersal of contaminants from the WIPP and also serve as a major route of exposure. The overall objective of this part of the WIPP EM is to determine whether the amounts and types of aerosols in the vicinity of the WIPP have been affected by activities at the site. A comprehensive summary of radiological aerosol studies is in preparation for release by May 2000. A complementary analysis of trace elements and aerosol ions has been conducted for the WIPP EM in support of the radionuclide studies and is reported herein.

The inorganic studies complement the radionuclide assessments because they provide information about the types of aerosols in the atmosphere and how their concentrations vary with time. In addition, some of the trace elements being studied (As, Ba, Cd, Cr, Pb, Hg, Se and Ag) are listed as components of the Permitted TRU Mixed Wastes in the WIPP hazardous waste permit (Waste Acceptance Criteria for the Waste Isolation Pilot Plant, DOE/WIPP-069, November 8, 1999). Since no mixed waste has yet been delivered to the site, the nonradiological data presented here can be considered part of a continuing effort to characterize background conditions.

### Methods

A detailed summary of the sampling design and analytical procedures for the WIPP EM aerosol studies was presented in the CEMRC 1998 Report and that material will not be repeated here. Briefly, ambient aerosol samples are collected from three sampling stations (On Site, Cactus Flats and Near Field). Aerosol samples for trace element (TE) and major ion analyses are collected using low-volume ( $10 \text{ L min}^{-1}$ ) TSP,  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  systems. The samples for analyses by ion chromatography (IC) are collected on 2- $\mu\text{m}$  pore-size, 47-mm

diameter Gelman Teflo® PTFE Teflon® filters (Gelman Sciences, Ann Arbor, MI) while the TE samples are collected on 0.8- $\mu\text{m}$  pore-size, 47-mm diameter cellulose-ester Gelman Metricel® filters. Since February 1998, TE/IC sampling periods of two, two and three days per week have been used (the filters are replaced on Monday, Wednesday and Friday). The analyses of the filters alternate between TE and IC, with every second sample archived (TE, archive, IC, archive, TE, archive, IC, archive etc.). The results presented here cover the periods 4 November 1997 – 29 June 1999 and 3 February 1998 – 29 July 1999 for the IC and TE analyses, respectively.

Gravimetric determinations of the aerosol mass collected on the filters were conducted at CEMRC, using standard operating procedures detailed in the 1998 CEMRC Report. These determinations were only made on the Teflo® (IC) filters because static problems with the Metricel® filters caused their weights to vary erratically. The total mass that accumulated during a sampling period was divided by the total air volume drawn through the filter to calculate aerosol mass concentrations.

All of the aerosol data presented in this report were produced by CEMRC. Standard operating procedures have been developed for the chemical analyses, and where possible these are based on applicable standard U.S. Environmental Protection Agency (EPA) procedures. A summary of the analytical procedures and quality control aspects of the chemical analyses of the aerosols and other WIPP EM samples are presented elsewhere in this report (Appendix K).

For the IC analyses, individual Teflo® filters were extracted with de-ionized water, using an ultrasonic water bath to facilitate the process. Aliquots of the same aqueous extracts of the aerosol samples were used for both anion and cation analyses.

Aerosol filters were prepared for elemental analyses using a microwave digestion system with  $\text{HNO}_3$ ,  $\text{HCl}$ ,  $\text{HF}$ ,  $\text{H}_2\text{O}_2$  and  $\text{H}_2\text{O}$ . The concentrations of major and trace elements

were determined in the aerosol samples by AAS, ICP-ES and ICP-MS. The ICP-MS was used in place of ICP-ES for analyses after January 1999.

## Results and Discussion

### Aerosol Mass Concentrations

The gravimetric data collected from the inception of the aerosol sampling program show that mass concentrations exhibit a seasonal cycle with peaks in concentrations typically occurring in spring and minima in late fall and early winter (Fig. 12). One of the fundamental objectives for the inorganic baseline studies is to characterize the temporal trends and especially the nature of differences between sites before mixed wastes are accepted at the WIPP. Only TSP data were available from the On Site station, and therefore the design used for comparing mass concentrations was to run two separate paired t-tests comparing the On Site station to the other two aerosol sampling stations. The strategy for these tests was to match the aerosol mass data by sampling date and directly compare the paired mass concentrations between sites. This strategy was employed as a result of the clear seasonal cycles observed in mass concentrations.

The paired t-tests showed that the mass concentrations at the On Site station were significantly different from those at either Near Field or Cactus Flats at  $p < 0.0001$ . The mean difference between paired samples from On Site vs. Near Field was  $6.17 \mu\text{g m}^{-3}$ ; thus the mean TSP mass loading at the On Site station was ~30% higher than that at Near Field (Fig. 13). The difference in mean mass concentrations between On Site and Cactus Flats was slightly smaller at  $5.16 \mu\text{g m}^{-3}$ , a difference of more than 25%.

The next step in the analysis was to compare the TSP mass concentrations at Near Field and Cactus Flats, again using a paired t-test. Results of this test showed that the aerosol mass concentrations at the two sites were not significantly different ( $p \gg 0.05$ ), indicating that the elevated TSP mass concentrations at the On Site station (presumably associated with WIPP site activities) did not affect Near Field.

The  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  mass concentrations for the 1997-1999 samples were quite comparable to a much smaller data set obtained with high volume samplers and summarized in the 1998 CEMRC Report and in Lee et. al. (1999, *J. Radioanalytical and Nuclear Chem.* 234, 267). As a follow-up on those studies, mass concentrations were compared among samplers for the much larger data set from the low-volume samples. For these tests a two-way analysis of variance (ANOVA) was used, with sampling site and particulate size sampler type (i.e., TSP,  $\text{PM}_{10}$  or  $\text{PM}_{2.5}$ ) as variable classes. Data for the On Site station could not be included in this test because only a TSP sampler was deployed at that station.

Results of the ANOVA for aerosol mass confirmed that the differences in mass concentrations between Near Field and Cactus Flats were not significant ( $p > 0.05$ ), and this test expanded the results to include the  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  data. Furthermore, the effect (if any) was small because the differences between the mean  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  mass concentrations at Near Field and Cactus Flats were small (< 1% and ~5%, respectively).

On the other hand, the ANOVA of the mass data stratified by site and particulate size showed that the differences in mass concentrations as a function of particulate size were highly significant ( $p < 0.001$ ). Further analysis showed that a bulk of the mass was carried on particles collected in the  $\text{PM}_{10}$  samples. Comparisons of the mean mass concentrations showed that the  $\text{PM}_{10}$  fraction accounted for  $\geq 87\%$  and  $\geq 73\%$  of the TSP mass at Near Field and Cactus Flats, respectively (Fig. 13). The  $\text{PM}_{2.5}$  fraction amounted to  $\geq 45\%$  of the TSP mass at Near Field and  $\geq 39\%$  at Cactus Flats.

The between-sites differences in these percentages suggest that the aerosol mass may have been somewhat higher in larger particles at Near Field compared with Cactus Flats. This mirrors studies of soil texture in the vicinity of each sampling station, reported herein (Surface soil Radionuclides and Inorganic Chemicals). Some additional studies of aerosol elemental size distributions are in progress in collaboration with Dr. Suilou Huang

(New Mexico Institute of Mining and Technology).

The issue of mass-particle size distributions is highly relevant to other studies undertaken for the WIPP EM because, as discussed below, the ionic and elemental composition of the aerosol particles varies strongly as a function of particle size. It is also possible, if not likely, that activity concentrations of the radionuclides of interest for the project also vary as a function of particle size, with submicrometer particles of particular importance (Bondietti, E. A., et. al., 1988, *J. Environ. Radioactivity* 6, 99) although large (>20  $\mu\text{m}$ ) radioactive particles also can be transported long distances (Pöllänen, R., et. al., 1988, *Atmos. Environ.* 31, 3575). In addition, one would expect that the atmospheric residence times of aerosols of different sizes would vary as a function of their dry deposition rates. For example, submicrometer particles would have relatively long residence times and would be particularly susceptible to long-range transport. Therefore, any compositional differences between sites for size-separated aerosols have important implications for any materials, including radionuclides, that are mainly removed from the atmosphere by dry deposition. The particle size information also could be related to the soil textural data discussed elsewhere in this report (Surface Soil Radionuclides and Inorganic Chemicals).

### **Aerosol Ions**

The finding of higher mass concentrations at the On Site station leads to the question of what substance or substances are responsible for the observed differences. One candidate for this is halite (NaCl) from the extensive salt deposits mined in the WIPP underground and stored on the surface of the site. Additionally, as shown elsewhere in this report (Radionuclides and Inorganics in WIPP Exhaust Air), the Na concentrations in the fixed air samples (FAS) from the WIPP exhaust shaft were enriched several hundred-fold over those at the WIPP EM aerosol sampling stations.

Despite the elevated salt concentrations in the FAS samples, the Na concentrations in TSP samples from the On Site station were not significantly different from those at Near Field or Cactus Flats (paired t-tests,  $p > 0.05$ , see

Fig. 14). It is worth noting that the Na determined through the IC analyses is water soluble Na, and this presumably includes little if any Na released from the matrices of insoluble mineral aerosol particles.

Further analyses showed that the Na-to-mass ratios did not differ among sites either. Therefore, salt did not make up as large a percentage of the aerosol mass at the On Site station as one might expect if salt were a disproportionately large contributor to the aerosol mass there. In fact, aerosol Na amounted to only 1-2% of the total mass at the sites (Table 4). If one assumes that the composition of the salt deposits is similar to that of sea water, one would multiply the Na concentration by 3.26 to estimate the mass of salt in the atmosphere, using the seawater composition of Millero (1996, *Chemical Oceanography* 2<sup>nd</sup> edition, CRC Press, Boca Raton, FL.). Based on this presumptive source material, the salt particles in the atmosphere would amount to < 10% of the aerosol mass, which is clearly not sufficient to account for the observed differences in mass concentrations among sites.

Aerosol nitrate and sulfate are other important atmospheric constituents, in large measure as a result of their roles in the biogeochemical cycling of nitrogen and sulfur. Aerosols composed of these ions also are of considerable interest to atmospheric scientists because of their light scattering properties and associated effects on the fluxes of solar radiation. Aerosol nitrate mainly originates from fossil fuel combustion (Singh, H. B., 1987, *Environ. Sci. and Tech.* 21, 320), including emissions from motor vehicles (Dignon, J., 1992, *Atmos. Environ.* 26A, 1157) while anthropogenic sulfate is mainly produced through the combustion of coal and residual fuel oil (Cullis, C. F. and M. M. Hirschler, 1980, *Atmos. Environ.* 14, 1263). Thus the concentrations of these major ions could be elevated at the WIPP by the normal operations of the physical plant (i.e. emissions from motor vehicles, generators and other combustion sources).

Somewhat surprisingly, the amount of aerosol sulfate at each of the sites was greater than the concentration of atmospheric salt aerosols (see Fig. 14). The mass of aerosol sulfate also was greater than that of nitrate,

with sulfate amounting to 10-15% of the total mass loading, and nitrate < 5% of the mass. However, neither sulfate nor nitrate differed significantly between the On Site and Near Field or Cactus Flats TSP samples (paired t-tests,  $p > 0.05$ ). Therefore, like salt, the combination of these two anions cannot account for the differences in aerosol mass concentrations among sites.

The actual contributions of the sulfate- and nitrate-containing aerosols to the mass concentrations cannot be determined more precisely because the cations ( $\text{Na}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{NH}_4^+$ , etc.) that are bound to the anions, have not been established. In fact, the cations would increase their contributions of these types of aerosols considerably. For example, if the sulfate aerosol were predominantly in the form of the mineral gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ), the increase in mass contribution of the aerosol would be nearly 80% relative to sulfate alone, but if the sulfate aerosol were in the form of anhydrite ( $\text{CaSO}_4$ ), the increase in mass contribution would be ~40%.

Based on the IC data obtained to date, the combination of salt and the major anions account for roughly 25% to 30% of the total TSP aerosol mass at On Site, assuming a 50% contribution of the cations associated with nitrate and sulfate. Again, an important point of these analyses with respect to the WIPP EM is that none of these substances differed significantly among sites, and their contributions to the aerosol mass concentrations cannot explain the observed higher TSP masses at On Site.

The mass differences between sites were further investigated using the cation data generated by IC analyses of the aqueous extracts of the Teflo® filters. These comparisons showed that the differences between sites in Ca and Mg concentrations were highly significant (ANOVA,  $p < 0.001$ ), and K concentrations were marginally significant ( $0.05 > p > 0.01$ ). Moreover, the concentrations of all of these cations were highest in the samples from the On Site station (Fig. 15), and all of these elements are strongly enriched with respect to the cation/Na ratios expected from the previously referenced sea salt composition of Milero (1996) (Table 5).

While Ca and Mg contributed to the higher mass concentrations at the On Site station,

they cannot, even in combination, account for more than ~40% of the observed differences in mass. For example, if one assumes that Ca exists as a 50%/50% mixture of ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ )/ $\text{CaCO}_3$ , which would not be unreasonable given the mass concentrations of sulfate, then the corresponding difference in masses between On Site and Near Field attributable to these Ca-bearing minerals would be  $2.6 \mu\text{g m}^{-3}$ . This is about 35% of the observed mass difference between the two sites (i.e.  $22.3$  minus  $15.2 \mu\text{g m}^{-3}$ , Fig. 13). The corresponding value for Mg, based on the same assumed anion mixture, would be much smaller, about 3%. These comparisons indicate that Ca- and Mg-bearing mineral aerosols contribute to the higher mass concentrations at the On Site station, but these minerals alone cannot explain the observed differences.

One might speculate that construction or maintenance activities, including any operations involving cement or road construction at the WIPP may have mobilized some Ca-containing particles, but we have no way to investigate this further with the existing data.

An independent and perhaps more compelling reason for monitoring these aerosol ions, Mg in particular, is that MgO will be used as a cap material in the underground. MgO is being stored in the underground, and preliminary analyses of the FAS data also indicate elevated concentrations of airborne Mg relative to those observed at the three aboveground aerosol monitoring stations. Therefore, it will be informative to monitor aerosol Mg concentrations to determine whether the differences between sites become more or less pronounced as the nature of the disposal operations changes over time.

### **Elemental Constituents**

The combined ICP-MS and AAS trace element analyses can provide information for up to thirty-nine elements, but several elements were below detection limits in all samples. Specifically, Sb, Se and Tl were below detection limits in all TSP samples from the On Site station. Of these, Se is of particular interest because this element is listed as a probable component of the mixed waste. The case of Ag is somewhat puzzling because this element was detected only once in the On Site

TSP samples compared with frequencies of ~20-30% in each of the six other sets of samples. Se, on the other hand, was rarely detected in any of the other groups of samples. It is also noteworthy that both Ag and Se are frequently detected in the FAS samples. A summary of the percentages of samples with selected trace element concentrations above detection limits is presented in Fig. 16. Interestingly, the patterns of elements detected at Cactus Flats were qualitatively different from those at Near Field and On Site, and the pattern was consistent across all types of samples. In particular, Ba and Pb were more commonly detected at On Site and Near Field while As, Cd, Hg and Se were detected in a higher percentage of samples from Cactus Flats.

The sources for atmospheric Ba are largely unstudied, and this element was not considered in either of the two seminal references on the sources of trace elements in the atmosphere (Pacyna, J. M., 1986. In Nriagu, J. O. and C. I. Davidson (eds.), *Toxic Metals in the Atmosphere*, John Wiley, NY; 2; Nriagu, J. O., 1989, *Nature* 338, 47). Ba is known to be a component of some lubricants, however. On the other end of the scale, the biogeochemical cycling of pollutant Pb has been extensively studied, largely as a result of the global contamination caused by the use of leaded gasoline and the related health effects (Patterson, C. C., and D. M. Settle, 1987, *Marine Chem.* 22, 137).

Gravimetric analyses were not performed on the cellulose ester filters used for the TE sampling because of problems obtaining stable weights, and therefore it is not possible to unequivocally determine which of the elements may have contributed to the higher mass concentrations at the On Site station relative to the other sites. However, a simple examination of the data shows that Ca has the highest concentration per unit volume air of all the elements determined, with an arithmetic mean of  $0.85 \mu\text{g m}^{-3}$ . This is somewhat lower than the average concentration determined by IC, but some of this difference may be due to the fact that the time periods encompassed by the two sets of measurements are slightly different as a result of the TE/IC sequence in sampling. Also, different types of filters were used for the IC and elemental analyses, and it

is possible that the filters differ somewhat in their particle collection efficiencies. Except for a few extreme instances in which the IC Ca concentrations were higher, the data produced by the two methods were in good agreement.

After Ca, the element exhibiting the next highest aerosol concentration is Al. The average Al concentrations were similar at all sites, ranging from  $0.405 \mu\text{g m}^{-3}$  at Cactus Flats to  $0.477 \mu\text{g m}^{-3}$  at Near Field, with On Site intermediate at  $0.442 \mu\text{g m}^{-3}$ . Al is commonly used as an indicator of mineral dust (e.g. Rahn, R. A., 1976, *The Chemical Composition of the Atmospheric Aerosol*, Tech. Report, Grad. Sch. of Oceanogr., Univ. Rhode Island, Kingston.), with average crustal material containing ~8% Al (Taylor, S. R. and S. M. McLennan, 1995, *Rev. of Geophys.* 33, 241), and therefore mineral matter is one of the dominant components of aerosol, amounting to ~25% of the total mass concentration On Site (based on the IC mass data) and ~30-40% at the other two aerosol sampling stations.

Differences in mineral aerosol concentrations between sites cannot explain the higher mass concentrations at the On Site station. The differences in mineral dust concentrations among sites were small, and in the case of On Site and Cactus Flats they were the reverse of the patterns for mass concentrations.

The final analysis of the elemental data was to compare the concentrations On Site to those at Near Field and Cactus Flats. A simple indicator was derived by dividing the mean concentration for each element for the On Site station by the corresponding means at the other two sites. This was done only for those elements with  $\geq 50\%$  of the samples above detection limits (Fig. 17). This index provides a simple measure of whether the concentrations were higher at On Site than the other sites, although the index does not lend itself to rigorous statistics.

Corroborating the previous interpretations from the IC data, the ICP-MS Ca and Mg concentrations at the On Site station were 30% to 70% higher than those at the other two sites. It is important to emphasize that these results are from a fully independent analysis on a separate set of samples, and therefore this parallel method provides a powerful means for

validating the results and conclusions from the IC analyses.

Sr exhibited the third highest concentration difference between On Site and the other sites. Even though the concentration of Sr was not high enough to account for the differences in mass concentrations, these elemental differences in aerosol composition do provide some information on what types of aerosols are most abundant at each site. Fig. 17 also shows that the On Site:Cactus Flats elemental ratios tended to be higher than the On Site:Near Field ratios, which is further evidence for compositional differences between Cactus Flats and Near Field.

The concentrations of the elements exhibiting high levels at On Site (relative to the other stations) were divided by the matching Al concentrations to provide a means for determining whether the elements of concern are mainly associated with mineral dust or are enriched relative to that source. For all of the elements that had high concentrations On Site, including Ca and Mg, the ratios to Al were higher than in the average crustal material composition of Taylor and McLennan (1995) (Table 6).

Another important use of the elemental data in the future will be for comparisons to radionuclide analyses. These comparisons will be particularly useful for U and Th. The ICP-MS analyses have generated a set of data for these two elements in aerosols, and these data will not only provide background information but also a basis for comparisons against other media.

The interpretation of the elemental data produced to date has provided some insight into the apparent perturbation of mass concentrations at the WIPP site, but much of the mass difference cannot be accounted for, suggesting a contribution by organic substances. Follow-up studies on elemental and organic carbon in aerosols are being considered for the upcoming year.

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

**Table 4. Percentage of Mass Contributed by Nitrate and Sulfate (without Cations) in Aerosol Samples Collected during November 1997 - June 1999**

Anion	Site	Type	<sup>a</sup> N	Median	<sup>b</sup> Mean	<sup>c</sup> SE
Nitrate	On Site	<sup>d</sup> TSP	49	2.9%	3.2%	0.3%
		<sup>e</sup> PM <sub>10</sub>	49	2.6%	2.6%	0.2%
	Near Field	<sup>f</sup> PM <sub>2.5</sub>	36	1.8%	1.9%	0.2%
		TSP	53	3.4%	4.2%	0.5%
	Cactus Flats	PM <sub>10</sub>	49	2.4%	2.7%	0.2%
		PM <sub>2.5</sub>	39	1.5%	1.6%	0.2%
		TSP	57	3.4%	3.6%	0.3%
Sulfate	On Site	TSP	52	8.5%	11.2%	1.2%
	Near Field	PM <sub>10</sub>	52	12.5%	15.3%	1.3%
		PM <sub>2.5</sub>	51	20.1%	22.1%	1.5%
		TSP	60	11.8%	13.7%	1.1%
	Cactus Flats	PM <sub>10</sub>	54	12.8%	15.3%	1.3%
		PM <sub>2.5</sub>	50	25.0%	23.8%	1.5%
		TSP	68	11.9%	12.8%	0.9%

<sup>a</sup>N = number of samples

<sup>b</sup>Mean = arithmetic mean

<sup>c</sup>SE = standard error

<sup>d</sup>TSP = total suspended particulate matter

<sup>e</sup>PM<sub>10</sub> = suspended particulate matter with aerodynamic diameter ≤ 10 μm

<sup>f</sup>PM<sub>2.5</sub> = suspended particulate matter with aerodynamic diameter ≤ 2.5 μm

**Table 5. Mean Cation:Na Ratios in Aerosol Samples Collected during November 1997 – June 1999**

Site	Type	K/Na	Mg/Na	Ca/Na
	<sup>a</sup> Seawater Reference	0.037	0.119	0.038
On Site	<sup>b</sup> TSP	0.86	0.69	8.96
Near Field	<sup>c</sup> PM <sub>10</sub>	0.79	0.31	2.87
	<sup>d</sup> PM <sub>2.5</sub>	1.62	0.32	1.29
	TSP	0.80	0.33	3.62
Cactus Flats	PM <sub>10</sub>	1.07	0.27	3.58
	PM <sub>2.5</sub>	0.63	0.25	2.33
	TSP	1.03	0.29	4.95

<sup>a</sup>Seawater reference composition from Millero, F. J., 1996, *Chemical Oceanography*, 2<sup>nd</sup> edition. p. 61., CRC Press, Boca Raton, FL.

<sup>b</sup>TSP = total suspended particulate matter

<sup>c</sup>PM<sub>10</sub> = suspended particulate matter with aerodynamic diameter ≤ 10 μm

<sup>d</sup>PM<sub>2.5</sub> = suspended particulate matter with aerodynamic diameter ≤ 2.5 μm

**Table 6. Mean Trace Element Ratios in Aerosol Samples Collected during November 1997 - June 1999**

Site	Type	Ba/Al (× 1000)	Ca/Al	Mg/Al	Pb/Al (× 1000)	Sr/Al (× 1000)
	<sup>a</sup> Crustal Reference	<b>6.84</b>	<b>0.037</b>	<b>0.17</b>	<b>0.249</b>	<b>4.35</b>
On Site	<sup>b</sup> TSP	14.1	1.97	0.35	2.22	7.5
Near Field	<sup>c</sup> PM <sub>10</sub>	13.4	1.32	0.25	3.97	6.0
	<sup>d</sup> PM <sub>2.5</sub>	13.7	1.55	0.22	4.09	8.0
Cactus Flats	TSP	11.8	1.32	0.26	3.46	5.5
	PM <sub>10</sub>	12.9	1.33	0.22	3.22	6.3
	PM <sub>2.5</sub>	14.5	1.50	0.24	4.36	7.4
	TSP	12.8	1.21	0.21	2.56	5.6

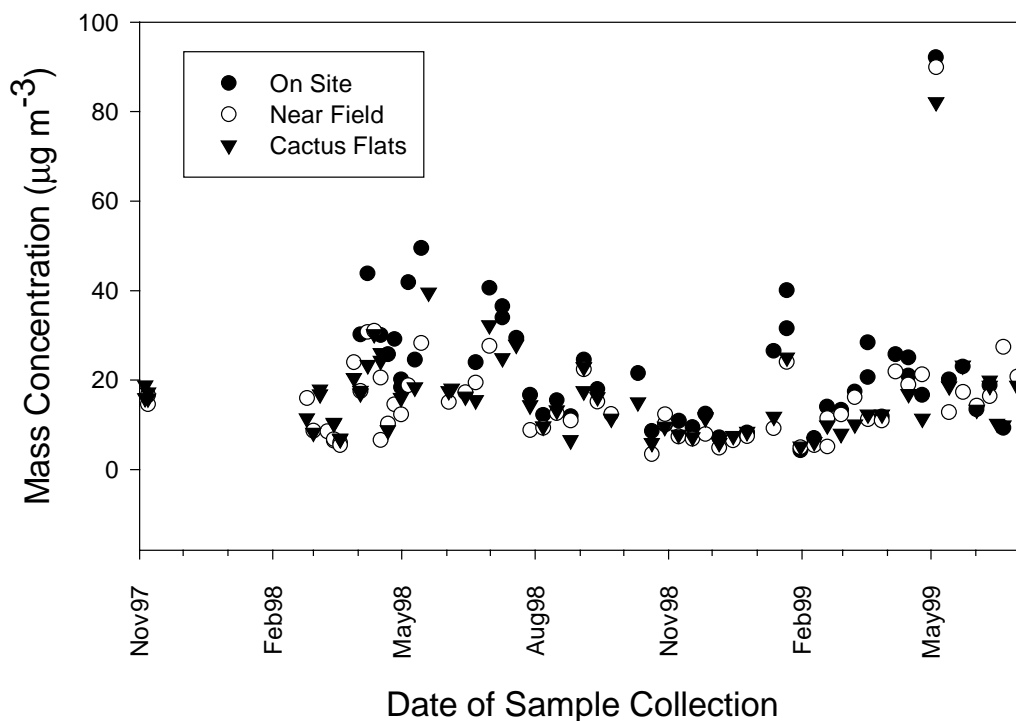
<sup>a</sup>Crustal reference composition from Taylor, S. R., and S. M. McLennan, 1995, *Rev. of Geophys.* 33, 241.

<sup>b</sup>TSP = total suspended particulate matter

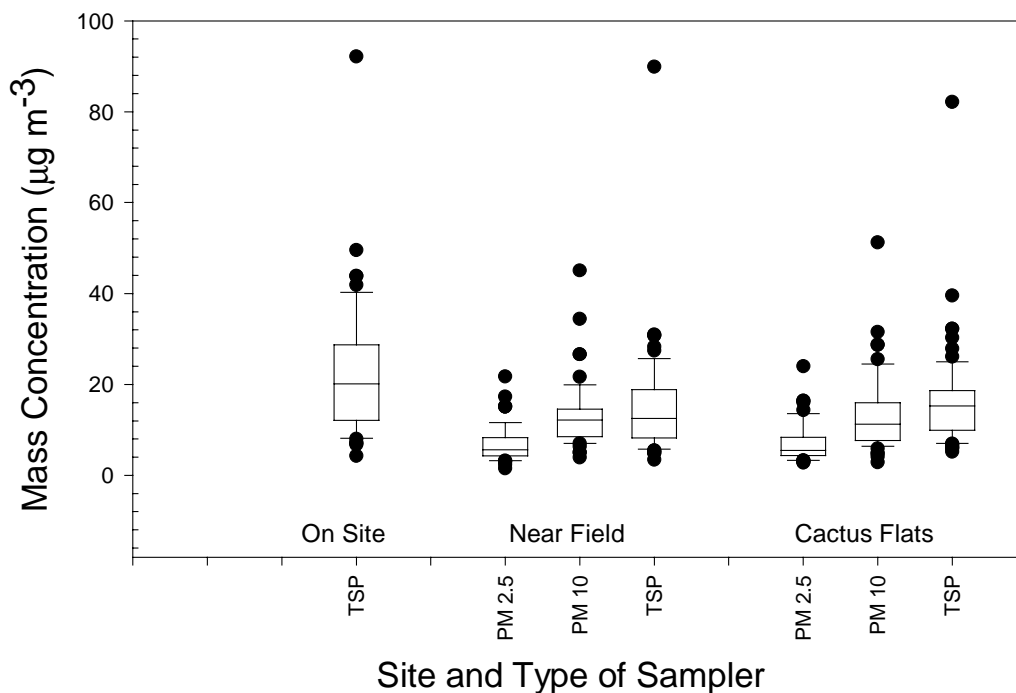
<sup>c</sup>PM<sub>10</sub> = suspended particulate matter with aerodynamic diameter ≤ 10 μm

<sup>d</sup>PM<sub>2.5</sub> = suspended particulate matter with aerodynamic diameter ≤ 2.5 μm



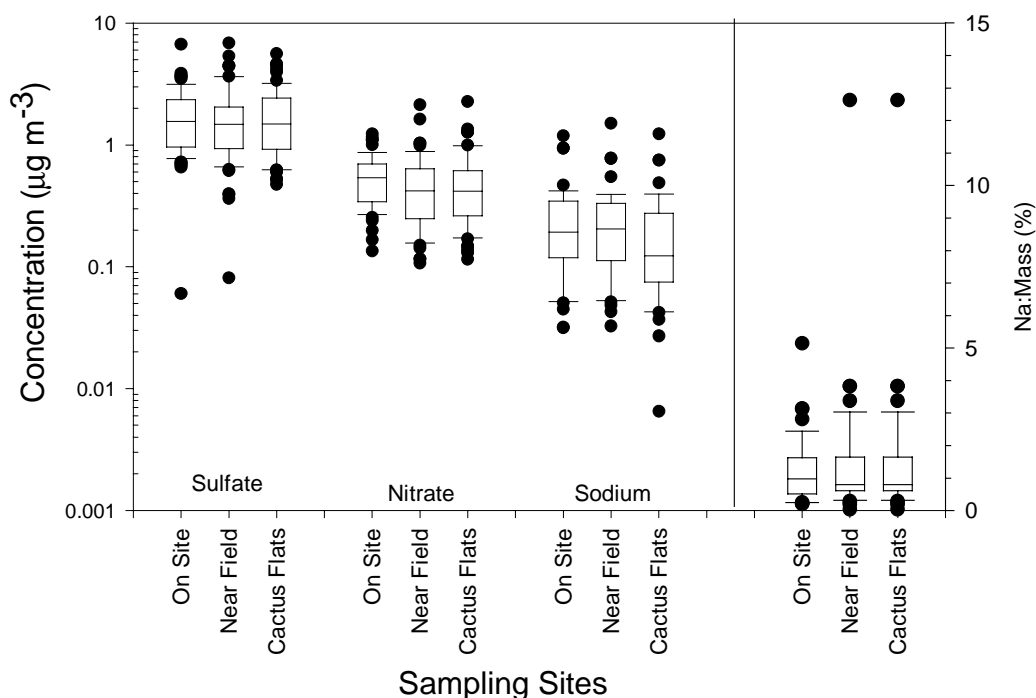


**Figure 12. Aerosol Mass Concentrations in TSP Samples Collected during November 1997 - June 1999**



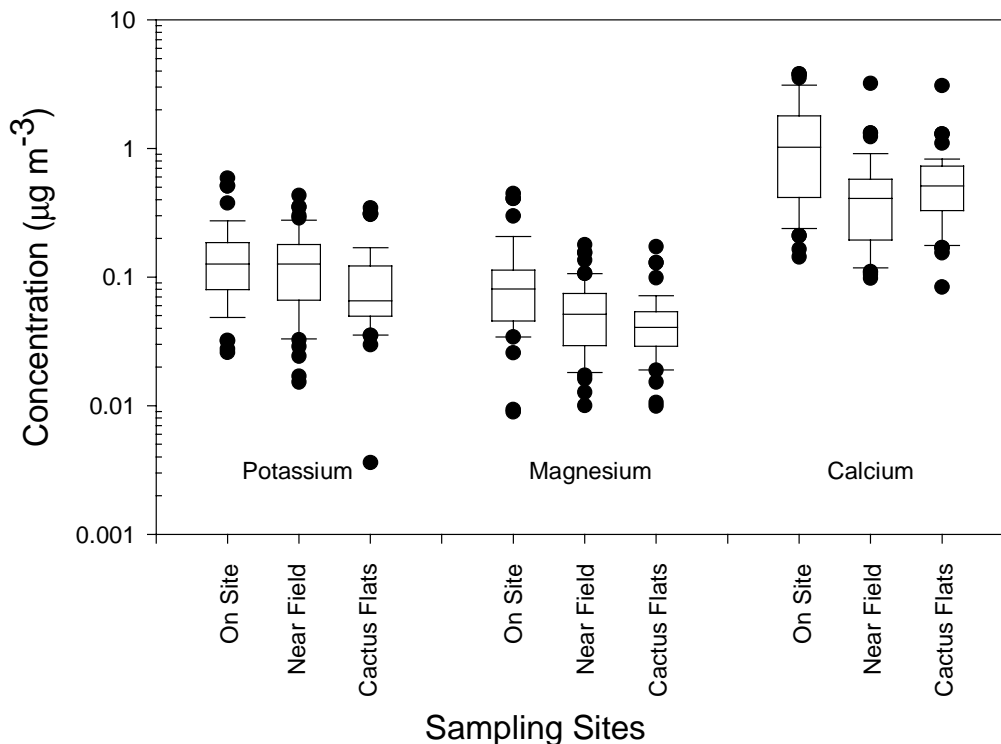
**Figure 13. Aerosol Mass Concentrations in Aerosol Samples Collected during November 1997 - June 1999**

The top and bottom of each box are the 25<sup>th</sup> and 75<sup>th</sup> percentiles, respectively. The line within each box is the median, and the external upper lines and lower lines are the 10<sup>th</sup> and 90<sup>th</sup> percentiles, respectively. Individual circles are values outside the 10<sup>th</sup> and 90<sup>th</sup> percentiles.



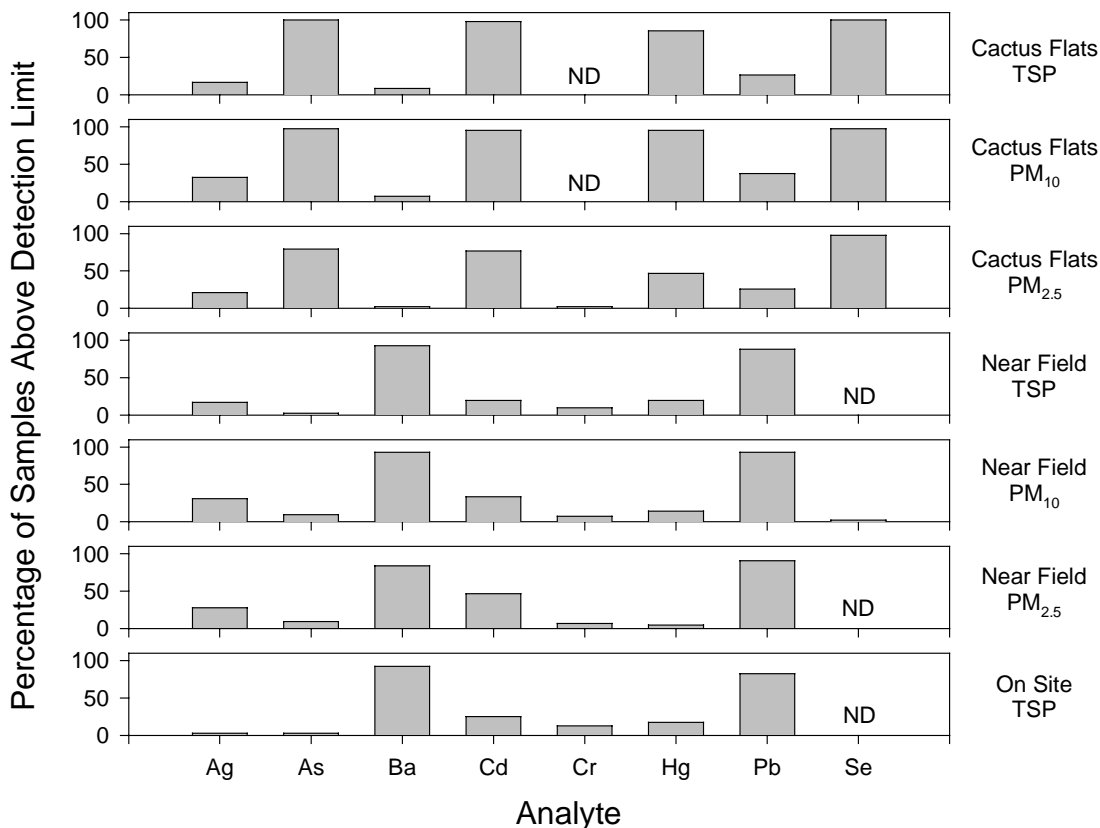
**Figure 14. Aerosol Ion Concentrations and Na:Mass Percentages in TSP Samples Collected during November 1997 - June 1999**

See explanation of box and whisker plots on Fig. 13.



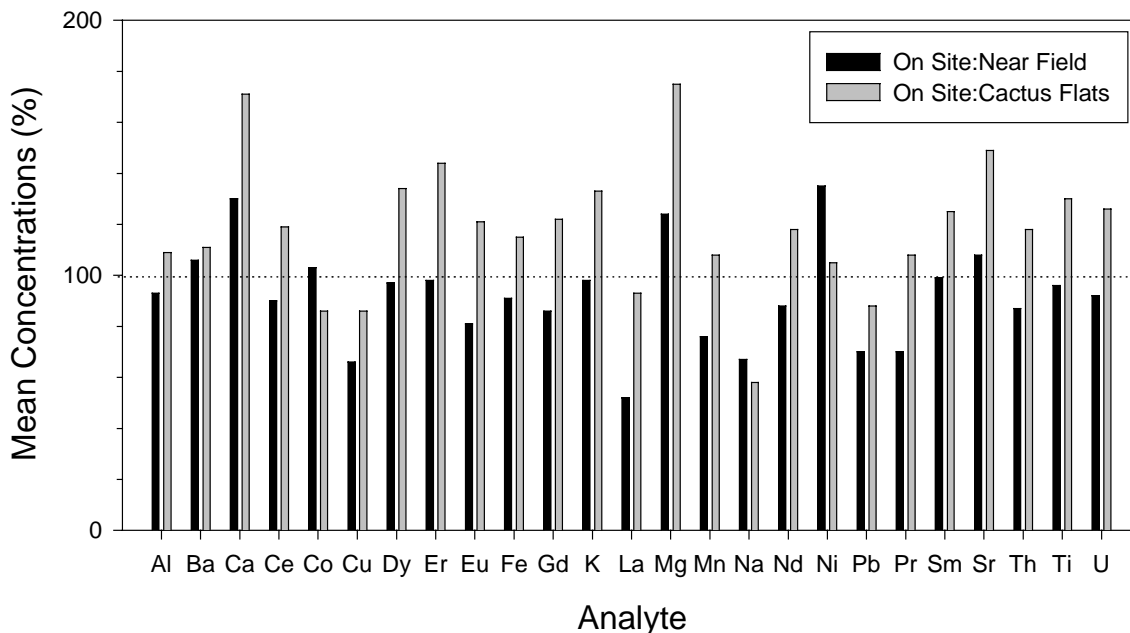
**Figure 15. Cation Concentrations in TSP Samples Collected during November 1997 - June 1999**

See explanation of box and whisker plots on Fig. 13.



**Figure 16. Percentages of Aerosol Samples with Selected Elemental Concentrations**

ND stands for not detected.



**Figure 17. Comparison of Mean Trace Element Concentrations at Three Aerosol Sampling Locations**

## Radionuclides and Inorganics in WIPP Exhaust Air

### Introduction

The CEMRC aerosol sampling program for the WIPP EM is designed to study the pathway that is the most likely route by which contaminants from the WIPP site could become rapidly dispersed in the environment. One facet of this comprehensive program is the monitoring of aerosols in the WIPP exhaust shaft. These samples are collected at a location (Station A) that represents the release point of aerosol effluents from the underground to the environment. CEMRC is interested in obtaining information on air quality from Station A because it provides a means for characterizing a source-term that will be needed for the interpretation of future monitoring results from the WIPP EM. For example, if radioactive or hazardous material was released from the WIPP, we would expect to detect it at Station A before it is observed in the local population or environment. In addition, source-term data collected at Station A would be of critical importance for the determination of public or worker dose in the event of an accident at the WIPP.

Another objective of the sampling program at Station A is to provide a gross check of emissions on a short resolution time-scale (e.g. weeks). For example, many of the WIPP EM analyses require many months to complete once the samples are collected. Such time is needed because of the specificity and sensitivity of the analyses. At Station A, gross monitoring results (less specific and sensitive) are provided within three weeks of sample collection and are used to trigger more detailed investigations if necessary.

### Methods

Samples are collected at Station A using a fixed air sampler (FAS). The FAS used by CEMRC was originally configured as an off-line continuous air monitor for aerosol radioactivity. However, the radiation detector of the sampler is no longer in operation and the sampler is operated as a FAS. The sampler is maintained and calibrated by Westing-

house/MK Ferguson, the maintenance and operations contractor for the WIPP site.

Aerosols are sampled in the exhaust shaft using a cylindrical shrouded probe (McFarland, A. R., et. al., 1989, *Environ. Sci. Technol.* 23, 1487; Ananda, N. K. and A. R. McFarland, 1989, *Am. Ind. Hyg. Assoc. J.* 50, 307). The shrouded probe is operated at 170 L min<sup>-1</sup> flow rate. At this flow rate, the shrouded probe has a transmission ratio (concentration measured by the probe relative to a reference concentration) of 0.93 to 1.11 for 10- $\mu$ m aerosols over the exhaust velocities expected at Station A (2 – 14 m s<sup>-1</sup>). Aerosols that enter the probe are then split into three FASs, each operating at 56 L min<sup>-1</sup>. Aerosol losses within the sampling system due to turbulent diffusion and gravitational settling are expected to be on the order of 13 to 30%. This system design was employed so that at least 50% of 10- $\mu$ m aerosols will be sampled under all exhaust-air flow rate conditions.

FAS samples are collected daily Monday through Friday (with the filter removed on Monday representing a three-day sample) using a 46-mm Versapore® filter. After sample collection, the filters are weighed, sealed and allowed to decay for approximately 7 days to eliminate any interference from naturally occurring, short-lived radionuclides.

The filters are individually screened for gross alpha/beta activity using a low background gas proportional counter with a count time of 24 hr. Calibration factors are used to correct for alpha and beta attenuation due to mass loading on individual filters. Typical values of minimum detectable concentration (MDC) for the gross alpha/beta analyses were 0.028 mBq m<sup>-3</sup> and 0.37 Bq g<sup>-1</sup> for gross alpha, and 0.073 mBq m<sup>-3</sup> and 1 Bq g<sup>-1</sup> for gross beta. Routine operational procedures include background, efficiency and alpha/beta cross-talk checks (Appendix L).

After the gross alpha/beta measurements, the five filters from each week (Monday - Friday) are digested and combined into a single weekly composite for elemental and gamma analyses. The filters are prepared using a

modification of EPA method 3052 for microwave digestion.

Ten mL of the digestate are sub-sampled for elemental constituent analysis. Elemental analyses for As, Se and Fe are performed by graphite furnace AA. All other analytes are determined by ICP-MS. Typical method detection limit (MDL) values (determined by analysis of matrix matched blanks and controls) for elemental analysis are provided in Table 7.

Following elemental analysis, an additional ten mL of the composite digestate are used for gamma-ray spectroscopy analyses. The gamma-ray analyses are performed using a low background, high purity Ge well detector and a count time of 48 hr. Typical values of MDC for gamma-ray emitting radionuclides are provided in Table 8. Routine operational procedures include gain stability, resolution, efficiency and background checks (Appendix L).

Upon completion of gamma-ray analyses, the sub-sample is recombined with the remaining digestate from that particular weekly composite. The weekly composites are stored until an approximate calendar year quarter is reached, and then combined into a single, quarterly composite. The quarterly composite is then analyzed for actinides using sequential separation and alpha spectrometry. The actinide separation and purification involves multiple precipitation, co-precipitation and ion exchange chromatography steps followed by precipitation and deposition with La. The efficiency for the actinide chemistry and spectrometry is determined from traced standards, and a traced matrix blank is analyzed with each composite. Routine operational procedures include background and gain stability checks (Appendix L)

Once verified and validated, analytical results are posted on the CEMRC web site (<http://www.cemrc.org>). Included on this web site are a program summary, time-series graphs and an analytical flow chart with photo illustrations. Gross alpha/beta, elemental and gamma data are reported within 21, 28 and 28 days, respectively, after the final filter of a week is collected (collected on a Monday after sampling through the weekend). Actinide data

are reported 42 days after the quarter is completed.

## Results and Discussion

Aerosol sampling has been conducted continuously at Station A by CEMRC since 12 December 1998. For measures of radioactivity, samples collected prior to receipt of waste at the WIPP (26 March 1999) were considered baseline. A single composite of the samples collected during the baseline was prepared for actinide analyses. The establishment of baseline for elemental constituents is on-going since WIPP has not received any mixed waste (containing both hazardous and radioactive constituents). For the purposes of this report, gross alpha/beta, elemental, gamma and actinide data are summarized through the end of second quarter 1999.

Values of gross alpha activity concentration and density ranged from 0.031 to 1.5 mBq m<sup>-3</sup> and 0.16 to 37 Bq g<sup>-1</sup>, respectively. Values of gross beta activity concentration and density ranged from 0.24 to 4.9 mBq m<sup>-3</sup> and 1.1 to 123 Bq g<sup>-1</sup>, respectively (Table 9). Interesting trends were observed in gross alpha/beta emissions (Figs. 18 and 19); in general, measured activity decreased after the WIPP began receiving waste (Table 11). This trend was most notable for gross alpha activity concentration, where measured activity for the 1999 second quarter was an order of magnitude lower than for the baseline. This trend may be the result of environmental phenomena, changes in WIPP operational practices or a combination of these factors. However, further investigation is necessary before any definitive conclusions can be drawn.

Numerous elemental constituents were observed in weekly composites (Table 10). Nearly 3/4 of all of the 39 elements were observed in 75-100% of the weekly composites. Tl and Sn were the only elements not detected in any weekly composite and these may serve as useful tracers for future WIPP aerosol studies. Many of the hazardous elements (e.g. Pb, Be, Cd, etc.) expected to be contained in WIPP mixed waste are already present in WIPP aerosol effluents. For many elements (Al, Ba, Ca, Co, Cu, Fe, K, Pb and V), volume concentrations were very similar to those reported in the CEMRC 1998 Report and herein

(Particulate Concentrations and Inorganics in near -Surface Air) for the TSP fraction at the On Site sampling location. Volume concentrations for Cr, Mg, Mn and Sr appear slightly enriched (> factor of 3 in concentration) relative to surface aerosols (as measured at the On Site location TSP). However, these differences may be attributed to changes in analytical methods, where data reported in the CEMRC 1998 Report were determined via ICP-emission spectrometry, not ICP-MS. A very significant enrichment (factor > 200) of Na was observed in effluent aerosols relative to surface aerosols. It is unlikely that such an increase for Na can be attributed to differences in analytical methods and is likely attributed to the presence of salt aerosols. Furthermore, this result was not surprising since salt is known to be a major component in WIPP effluents. The described enrichments, validated in subsequent analyses, may prove to be useful in determining the source of WIPP effluent aerosols.

The elemental constituent analyses confirm the presence of U and Th in WIPP effluent aerosols, which produce some portion of the gross alpha/beta signal. Weekly U and Th concentrations do not show a trend similar to that of gross alpha activity concentration (Fig. 20), suggesting that other alpha- and beta-emitting radionuclides influence the trends in gross alpha and beta.

With the exception of  $^7\text{Be}$ , no gamma-emitting radionuclides were routinely measured during baseline or second quarter samples. It is somewhat surprising that other naturally occurring radionuclides (e.g.  $^{40}\text{K}$ ) were not detected at a higher frequency. However, the sensitivity of gamma-ray spectroscopy is poor for the small aliquot used for these analyses.

$^7\text{Be}$  was detected in approximately 31% of samples, ranging in activity concentration and density from 5.3 to 12 mBq  $\text{m}^{-3}$  and 41 to 187 Bq  $\text{g}^{-1}$ , respectively. For detectable results, mean values ( $\pm\text{SE}$ ) of activity concentration and density were 8.4 ( $\pm 0.96$ ) mBq  $\text{m}^{-3}$  and 124 ( $\pm 15$ ) Bq  $\text{g}^{-1}$ , respectively. These results indicate that the aerosols entering through the WIPP air intake eventually reach the exhaust system and are released as exhaust effluents. The presence of  $^7\text{Be}$  in the exhaust

is an indicator of this mechanism because  $^7\text{Be}$  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). This finding may be of importance because other aerosols containing radionuclides of concern (e.g. Pu,  $^{137}\text{Cs}$ ) may follow a similar process and be detected in the exhaust in the absence of a WIPP-related contamination event. Therefore,  $^7\text{Be}$  may be a useful tracer for understanding aerosol residence times in the WIPP.

Naturally occurring U and Th isotopes were detected in the baseline and second quarter composites (Table 11). For the baseline composite,  $^{239,240}\text{Pu}$ ,  $^{238}\text{Pu}$  and  $^{241}\text{Am}$  results were less than MDC for the baseline composite. Second quarter results for  $^{238}\text{Pu}$  were also less than MDC.

Second quarter results for  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  were invalidated due to contamination discovered in the CEMRC gravimetrics laboratory. An exempt quantity source used for static control in the gravimetric process was found to have been damaged, contaminating a balance pan with low levels of  $^{241}\text{Am}$  and trace amounts of  $^{239,240}\text{Pu}$ . The contamination level was such that it was not observed through the routine gross alpha / beta or gamma measurements described herein, but was measured in the second quarter composite. No contamination was observed in the baseline composite. Although not reported herein, the 1999 third quarter composite was similarly affected, since the problem was not discovered until late third quarter. All decontamination and corrective actions were completed and verified to ensure that future results are not affected and similar situations are avoided. No other results were affected, because the affected balance is not used for weighing any other samples that are analyzed for Pu and Am. It is also important to note that second quarter and third quarter gross alpha results may be biased high due to contamination of some filters.

$^{234}\text{U}$  results were indistinguishable (when considered with two standard deviations) from those of  $^{238}\text{U}$  for activity concentration and density, suggesting secular equilibrium between the two isotopes (Table 11). Such results are expected for many natural sources of

U. At two standard deviations,  $^{228}\text{Th}$  activity concentration and density appeared to be enriched by  $\cong 2$  in comparison to  $^{232}\text{Th}$ . This disequilibrium, if validated as a continuing phenomenon, may be the result of the presence of the more mobile, intermediate element,  $^{228}\text{Ra}$ .

Activity densities of U and Th isotopes in WIPP effluent aerosols are very similar to those reported in the CEMRC 1998 Report and herein for soils collected in the vicinity of the WIPP (Surface Soil Radionuclides and Inorganic Chemicals). U and Th isotope concentrations are virtually identical between the baseline and second quarter composites, again

suggesting that other alpha- and beta- emitting radionuclides are influencing the trends observed in the gross alpha/beta data. Since active mining operations in the WIPP underground resumed in 1999, the FAS used by CEMRC has experienced extensive air flow reductions, resulting in failure to maintain the instrument's minimum specifications. Measures to eliminate this source of bias in the sampling are currently under development. Tables presenting aerosol data summarized herein are available on the CEMRC web site at <http://www.cemrc.org>.

**Table 7. Typical MDL Values for Elemental Analysis of Weekly Composites of FAS Samples**

Element	Typical <sup>a</sup> MDL		Element	Typical <sup>a</sup> MDL	
	Volume Concentration (ng m <sup>-3</sup> )	Mass Concentration (ng mg <sup>-1</sup> )		Volume Concentration (ng m <sup>-3</sup> )	Mass Concentration (ng mg <sup>-1</sup> )
Ag	1.7E-02	2.6E-01	Li	5.7E-01	8.7E+00
Al	4.9E+01	7.5E+02	Mg	1.3E-01	2.0E-02
As	3.7E-01	5.7E+00	Mn	4.7E-01	7.1E+00
Ba	8.7E-01	1.3E+01	Mo	9.6E-01	1.5E+01
Be	8.5E-02	1.3E+00	Na	1.0E+03	1.4E+04
Ca	1.3E+02	2.0E+03	Nd	8.0E-03	1.2E-01
Cd	9.5E-02	1.5E+00	Ni	2.1E+00	3.2E+01
Ce	1.1E-02	1.6E-01	Pb	1.1E-01	1.7E+00
Co	3.7E-01	5.7E+00	Pr	4.0E-03	6.1E-02
Cr	1.1E+01	1.6E+02	Sb	8.9E-01	1.4E+01
Cu	8.7E-01	1.3E+01	Se	3.2E-01	4.9E+00
Dy	2.7E-03	4.0E-02	Sm	4.0E-03	6.1E-02
Er	4.0E-03	6.1E-02	Sn	4.8E+01	7.3E+02
Eu	2.7E-03	4.0E-02	Sr	8.0E-01	1.2E+01
Fe	7.7E+00	1.2E+02	Th	5.3E-03	8.1E-02
Gd	2.7E-03	4.0E-02	Ti	6.3E+00	9.5E+01
Hg	2.7E-02	4.0E-01	Tl	1.6E+01	2.4E+02
K	7.1E+01	1.1E+03	U	4.0E-03	6.1E-02
La	8.0E-03	1.2E-01	V	1.2E+00	1.8E+01
Li	5.7E-01	8.7E+00	Zn	4.6E+01	5.4E+02
Mg	1.3E-01	2.0E-02			
Mn	4.7E-01	7.1E+00			
Mo	9.6E-01	1.5E+01			

<sup>a</sup>MDL = method detection limit

**Table 8. Typical MDC Values for Gamma-Ray Emitting Radionuclides Analyzed in Weekly Composites of FAS Samples**

Radionuclide	Typical <sup>a</sup> MDC		Radionuclide	Typical <sup>a</sup> MDC	
	Activity Concentration (Bq m <sup>-3</sup> )	Activity Density (Bq g <sup>-1</sup> )		Activity Concentration (Bq m <sup>-3</sup> )	Activity Density (Bq g <sup>-1</sup> )
<sup>228</sup> Ac	2.4E-03	3.4E+01	<sup>40</sup> K	6.3E-03	9.0E+01
<sup>241</sup> Am	3.4E-04	4.7E+00	<sup>54</sup> Mn	4.2E-04	5.9E+00
<sup>133</sup> Ba	2.8E-04	4.0E+00	<sup>237</sup> Np	8.5E-04	1.2E+01
<sup>140</sup> Ba	6.2E-01	5.0E+03	<sup>234m</sup> Pa	6.3E-02	9.1E+02
<sup>7</sup> Be	9.6E-03	1.1E+02	<sup>210</sup> Pb	3.7E-03	5.4E+01
<sup>212</sup> Bi	3.8E-03	5.6E+01	<sup>212</sup> Pb	7.3E-04	1.1E+01
<sup>214</sup> Bi	1.5E-03	2.1E+01	<sup>214</sup> Pb	1.1E-03	1.5E+01
<sup>141</sup> Ce	2.0E-03	2.0E+01	<sup>(b)iso</sup> Pu	4.2E-02	6.1E+02
<sup>144</sup> Ce	1.4E-03	1.9E+01	<sup>(c)226</sup> Ra	5.4E-03	7.5E+01
<sup>252</sup> Cf	4.9E-02	7.0E+02	<sup>103</sup> Ru	1.9E-03	2.1E+01
<sup>244</sup> Cm	4.1E-02	5.9E+02	<sup>106</sup> Ru	4.5E-03	6.2E+01
<sup>58</sup> Co	8.8E-04	1.1E+01	<sup>125</sup> Sb	1.2E-03	1.7E+01
<sup>60</sup> Co	8.6E-04	1.2E+01	<sup>228</sup> Th	9.2E-03	1.4E+02
<sup>51</sup> Cr	2.4E-02	2.4E+02	<sup>232</sup> Th	6.2E-02	8.6E+02
<sup>134</sup> Cs	4.7E-04	6.7E+00	<sup>234</sup> Th	1.8E-03	2.6E+01
<sup>137</sup> Cs	5.2E-04	7.6E+00	<sup>208</sup> Tl	5.9E-04	8.5E+00
<sup>152</sup> Eu	9.0E-04	1.3E+01	<sup>233</sup> U	2.0E-03	2.8E+01
<sup>154</sup> Eu	1.0E-03	1.5E+01	<sup>235</sup> U	1.1E-03	1.5E+01
<sup>155</sup> Eu	5.2E-04	7.4E+00	<sup>88</sup> Y	1.6E-03	2.1E+01
<sup>59</sup> Fe	4.2E-03	4.7E+01	<sup>65</sup> Zn	1.4E-03	1.9E+01
<sup>131</sup> I	1.1E+01	8.3E+04	<sup>95</sup> Zr	1.7E-03	2.1E+01
<sup>192</sup> Ir	5.2E-04	6.4E+00			

<sup>a</sup>MDC = minimum detectable concentration

<sup>b</sup>Plutonium isotopes cannot be distinguished by the analytical method; the values of MDC are based on the x-ray yield of <sup>238</sup>Pu.

<sup>c</sup>Gamma rays from <sup>235</sup>U will interfere with the measurement of <sup>226</sup>Ra; the values of MDC are based on the absence of <sup>235</sup>U in the sample



**Table 9. Summary Statistics for Gross Alpha/Beta Analyses of Daily FAS Filters**

Gross Emission	Activity Concentration (Bq m <sup>-3</sup> )			Activity Density (Bq g <sup>-1</sup> )		
	<sup>a</sup> N	<sup>b</sup> Mean	<sup>c</sup> SE	N	Mean	SE
<b>Pre-Operational Baseline</b>						
Alpha	71	3.1E-04	3.1E-05	71	3.6E+00	5.8E-01
Beta	71	1.1E-03	9.1E-05	71	1.4E+01	1.9E+00
<b>Operational Monitoring Second Quarter, 1999</b>						
Alpha	65	1.1E-04	6.5E-06	65	1.7E+00	1.6E-01
Beta	65	8.2E-04	2.3E-05	65	1.6E+01	1.6E+00

<sup>a</sup>N = number of samples included in calculations<sup>b</sup>Mean = arithmetic mean<sup>c</sup>SE = standard error of mean

**Table 10. Summary Statistics for Elemental Constituents in Weekly FAS Composites Collected during 12 December 1998 – 30 June 1999**

Element	<sup>a</sup> Frequency of Detection (%)	Volume Concentration (ng m <sup>-3</sup> )			Mass Concentration (ng mg <sup>-1</sup> )		
		N	Mean	SE	N	Mean	SE
Ag	90	26	8.1E-02	1.0E-02	26	1.1E+00	1.2E-01
Al	100	29	8.6E+02	2.5E+02	29	1.0E+04	2.9E+03
As	86	25	1.7E+00	2.5E-01	25	1.9E+01	2.4E+00
Ba	100	29	8.3E+00	8.2E-01	29	1.0E+02	9.0E+00
Be	14	4	3.6E-01	2.1E-01	4	4.1E+00	2.4E+00
Ca	100	29	1.9E+03	2.3E+02	29	2.3E+04	2.3E+03
Cd	93	27	5.3E-01	8.7E-02	27	7.1E+00	1.2E+00
Ce	100	29	8.4E-01	9.2E-02	29	1.0E+01	8.7E-01
Co	93	27	1.3E+00	1.5E-01	27	1.6E+01	2.3E+00
Cr	21	6	1.8E+01	2.0E+00	6	1.4E+02	2.5E+01
Cu	100	29	3.5E+01	2.8E+00	29	4.6E+02	3.3E+01
Dy	100	29	5.4E-02	7.0E-03	29	6.6E-01	7.2E-02
Er	100	29	3.3E-02	5.3E-03	29	4.0E-01	5.7E-02
Eu	97	28	1.5E-02	1.6E-03	28	1.7E-01	1.6E-02
Fe	100	29	1.0E+03	9.5E+01	29	1.3E+04	1.1E+03
Gd	100	29	7.7E-02	1.1E-02	29	9.6E-01	1.3E-01
Hg	24	7	6.2E-02	1.3E-02	7	5.9E-01	1.3E-01
K	97	28	8.9E+02	1.0E+02	28	9.8E+03	7.0E+02
La	100	29	4.8E-01	5.6E-02	29	5.8E+00	5.1E-01
Li	83	24	1.5E+00	1.8E-01	24	1.6E+01	1.2E+00
Mg	100	29	1.1E+03	1.3E+02	29	1.2E+04	8.2E+02
Mn	100	29	5.3E+01	7.6E+00	29	7.1E+02	1.2E+02
Mo	48	14	1.9E+00	2.5E-01	14	2.0E+01	3.0E+00
Na	97	28	4.1E+04	8.9E+03	28	4.5E+05	8.9E+04
Nd	100	29	3.6E-01	4.0E-02	29	4.4E+00	3.9E-01
Ni	83	24	5.7E+00	6.9E-01	24	6.9E+01	6.7E+00
Pb	100	29	6.4E+00	9.9E-01	29	8.0E+01	9.4E+00
Pr	100	29	1.0E-01	1.1E-02	29	1.2E+00	1.0E-01
Sb	100	29	4.2E+01	4.6E+00	29	6.2E+02	8.9E+01
Se	24	7	4.8E-01	6.3E-02	7	8.4E+00	1.3E+00
Sm	100	29	3.9E-01	2.5E-02	29	5.4E+00	4.7E-01
Sn	0	0	<sup>b</sup> NA	NA	NA	NA	NA
Sr	100	29	2.6E+01	3.1E+00	29	2.9E+02	1.9E+01
Th	100	29	1.3E-01	1.6E-02	29	1.6E+00	1.6E-01
Ti	97	28	4.0E+01	4.3E+00	28	5.0E+02	4.9E+01
Tl	0	0	NA	NA	NA	NA	NA
U	76	22	6.0E-02	1.2E-02	22	7.4E-01	1.4E-01
V	55	16	2.4E+00	2.9E-01	16	2.9E+01	3.5E+00
Zn	93	27	4.4E+02	1.8E+02	27	5.4E+03	2.1E+03

<sup>a</sup>Percent of samples with values greater than detection limit<sup>b</sup>NA = not applicable

**Table 11. Results of Actinide Analyses for Baseline and Second Quarter 1999 FAS Composite Samples**

Radionuclide	Activity Concentration (Bq m <sup>-3</sup> )			Activity Density (Bq g <sup>-1</sup> )		
	<sup>a</sup> C	<sup>b</sup> SD	<sup>c</sup> MDC	C	SD	MDC
<b>Preoperational Baseline Composite (26 December 1998 – 25 March 1999)</b>						
<sup>238</sup> Pu	<MDC	<sup>d</sup> NA	2.4E-08	<MDC	N/A	3.00E-04
<sup>239</sup> Pu	<MDC	NA	2.4E-08	<MDC	NA	2.87E-04
<sup>241</sup> Am	<MDC	NA	5.5E-08	<MDC	NA	6.90E-04
<sup>228</sup> Th	7.6E-07	5.2E-08	9.7E-08	8.1E-03	5.6E-04	1.23E-03
<sup>230</sup> Th	7.0E-07	4.9E-08	6.8E-08	7.5E-03	5.3E-04	8.33E-04
<sup>232</sup> Th	4.9E-07	3.7E-08	3.6E-08	5.2E-03	4.0E-04	4.29E-04
<sup>234</sup> U	8.9E-07	4.9E-08	3.0E-08	9.5E-03	5.3E-04	3.83E-04
<sup>235</sup> U	4.1E-08	1.5E-08	2.7E-08	4.4E-04	1.6E-04	3.17E-04
<sup>238</sup> U	8.5E-07	4.9E-08	2.4E-08	9.1E-03	5.2E-04	3.00E-04
<b>Operational Monitoring Composite (26 March 1999 – 30 June 1999)</b>						
<sup>238</sup> Pu	<MDC	NA	2.4E-08	<MDC	NA	3.00E-04
<sup>239</sup> Pu	<sup>e</sup> NR	NR	2.4E-08	NR	NR	2.87E-04
<sup>241</sup> Am	NR	NR	5.5E-08	NR	NR	6.90E-04
<sup>228</sup> Th	1.1E-06	7.0E-08	9.7E-08	1.5E-02	9.6E-04	1.23E-03
<sup>230</sup> Th	5.6E-07	4.6E-08	6.8E-08	7.6E-03	6.3E-04	8.33E-04
<sup>232</sup> Th	5.8E-07	4.0E-08	3.6E-08	7.9E-03	5.5E-04	4.29E-04
<sup>234</sup> U	7.3E-07	4.6E-08	3.0E-08	9.9E-03	6.2E-04	3.83E-04
<sup>235</sup> U	3.3E-08	1.2E-08	2.7E-08	4.5E-04	1.6E-04	3.17E-04
<sup>238</sup> U	6.1E-07	4.1E-08	2.4E-08	8.4E-03	5.6E-04	3.00E-04

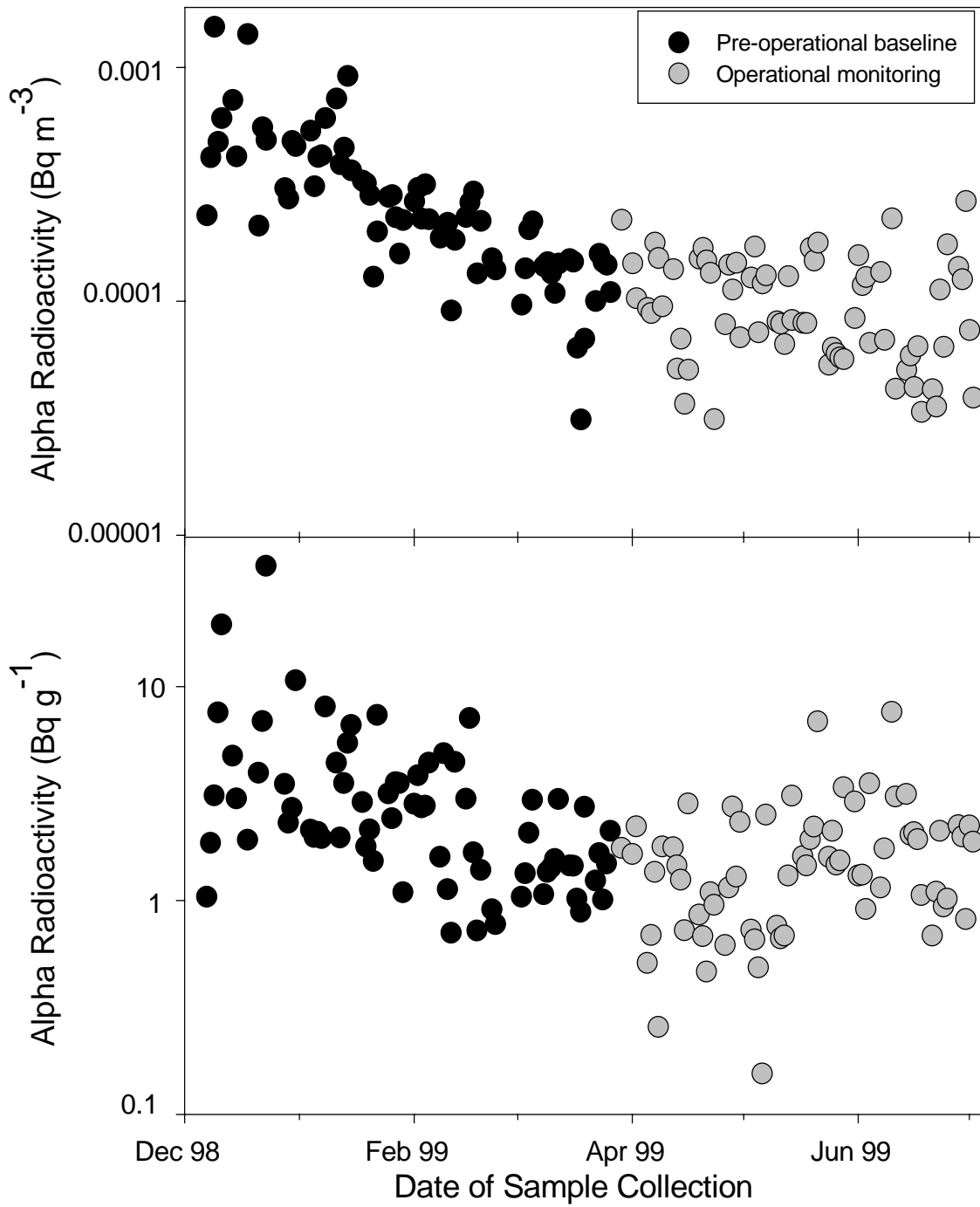
<sup>a</sup>C = activity concentration

<sup>b</sup>SD = standard deviation

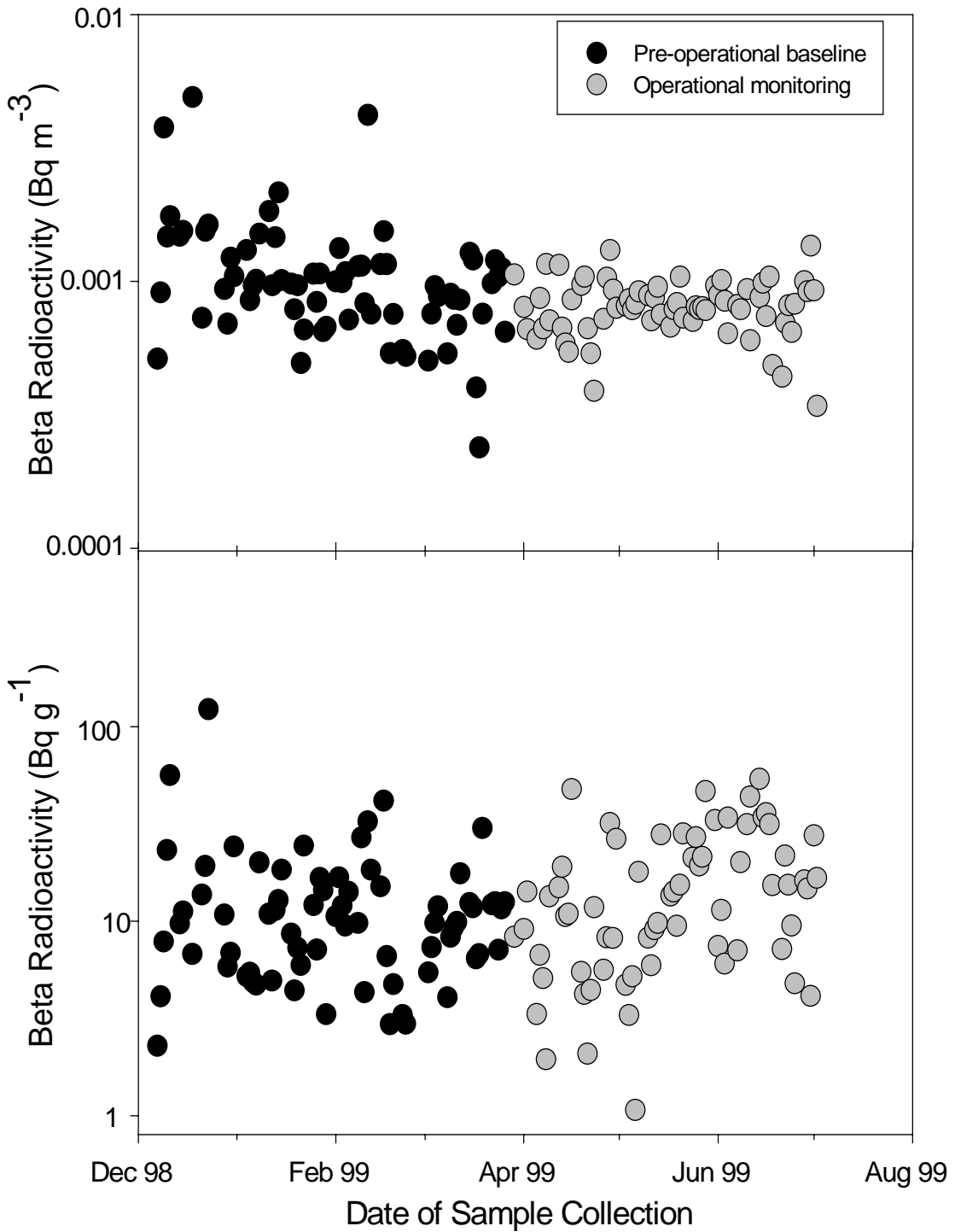
<sup>c</sup>MDC = minimum detectable concentration.

<sup>d</sup>NA = not applicable

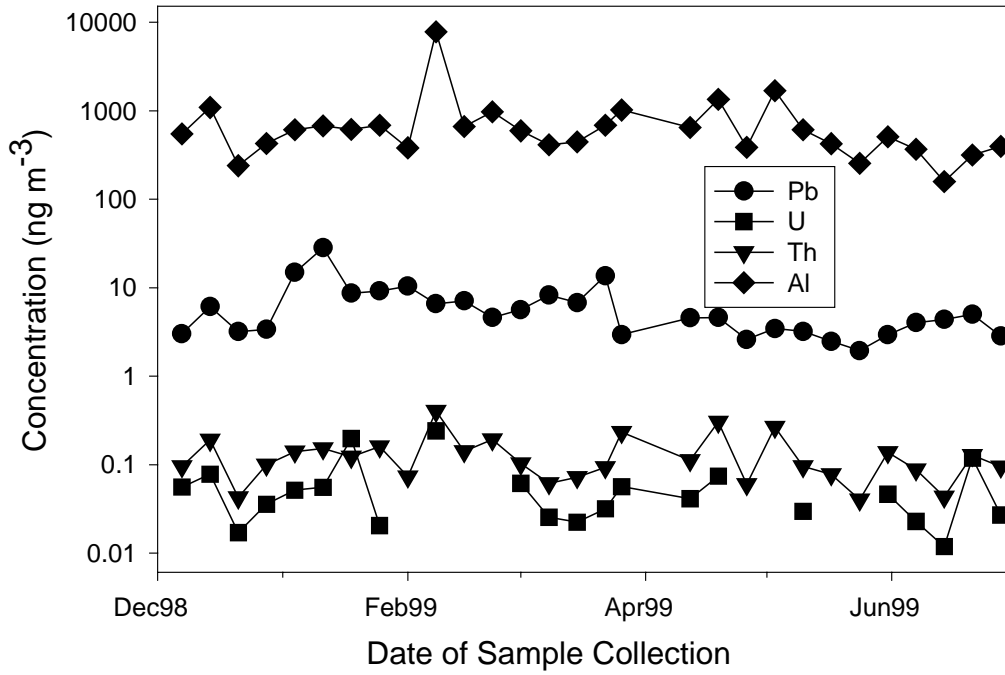
<sup>e</sup>NR = not reported



**Figure 18. Alpha Emitting Radioactivity in FAS Samples Collected during December 1998 – July 1999**



**Figure 19. Beta Emitting Radioactivity in FAS Samples Collected during December 1998 – July 1999**



**Figure 20. Selected Elemental Constituents Released as Aerosols in the Exhaust from the WIPP**

## Surface Soil Radionuclides and Inorganic Chemicals

### Introduction

Results reported herein are from soil samples collected during 1998 from a grid of 16 locations surrounding the WIPP site (the Near Field grid) and a grid of 16 locations approximately 12 miles southeast of the WIPP (the Cactus Flats grid, Fig. 2). Radioanalyses of a subset of these samples for  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{228}\text{Th}$  and  $^{239,240}\text{Pu}$  were reported in the CEMRC 1998 Report using measurements made by Duke Engineering and Services (DES) (Bolton, Massachusetts). Measurements presented herein were made by CEMRC for those same radio-analytes plus  $^{137}\text{Cs}$ ,  $^{208}\text{Tl}$ ,  $^{212}\text{Pb}$ ,  $^{212}\text{Bi}$ ,  $^{214}\text{Pb}$ ,  $^{228}\text{Ac}$ ,  $^{234\text{m}}\text{Pa}$ ,  $^{241}\text{Am}$ ,  $^{40}\text{K}$ ,  $^{60}\text{Co}$  and  $^7\text{Be}$ . The natural radionuclides  $^{208}\text{Tl}$ ,  $^{212}\text{Bi}$ ,  $^{214}\text{Pb}$  and  $^{212}\text{Pb}$  are measured after allowing for ingrowth and can be used to estimate the concentration of Ra. However, these measurements do not reflect natural levels of those radionuclides in the environment. Results are also presented for 36 non-radiological analytes measured using ICP-MS, AAS and IC.

One finding presented in the CEMRC 1998 Report was that there were significant differences in many analyte concentrations between the Near Field and Cactus Flats grids. Differences in soil texture were postulated as a possible cause for these differences. Aliquots of 73 of the 96 samples collected in 1998 were subsequently analyzed for soil texture in order to test this hypothesis.

### Methods

The 16 sampling locations comprising each grid are distributed over approximately 16,580 hectares. At each of the 32 locations, soil was collected at three randomly selected sites within a 50-m radius of the selected reference point. Individual sampling sites were selected on the basis of: relatively flat topography, minimum surface erosion and minimum surface disturbance by human or livestock activity. At each sampling site, approximately 12 L of soil were collected from within two 50-cm x 50-cm areas, to a depth of approximately 2 cm. Soil samples were excavated using a

trowel and placed in plastic bags for transport and storage. Sampling equipment was cleaned between samples.

Initial preparation of the samples consisted of passing the soil through a 2-mm sieve to remove rocks, roots and other materials. One half of the 96 samples were originally analyzed for inorganic analytes in 1998. These soil samples were homogenized in the laboratory using a riffler. The method of homogenization was shown previously to yield subsamples that differed from the overall mean count of a radioactive tracer ( $^{137}\text{Cs}$ ) by no more than 7%.

Each sample was homogenized again before aliquots were split for inorganic/metal analyses. New aliquots obtained from samples analyzed and reported in the 1998 CEMRC Report had already been dried at 105° C. Therefore, no analysis for Hg was performed on this set of 48 samples. A 50-g aliquot was removed for radiochemical analyses and ~300 mL aliquots were used for gamma spectroscopy analysis. The samples for gamma analysis were sealed in a ~300 mL paint can and stored for at least 21 days to allow radon progeny to reach equilibrium with parent radionuclides.

Soil samples were analyzed by AAS for As and Se. ICP-MS was used to analyze samples for Ag, Al, As, Ba, Be, Ca, Cd, Co, Cr, Cu, Eu, Fe, Hg, K, La, Li, Mg, Mn, Mo, Na, Ni, Pb, Sb, Sr, Th, Ti, Tl, U, V and Zn. The lower detection limits for both of these systems are in the low parts per billion range (Appendix K). Soil samples were analyzed by IC for chloride, nitrate, nitrite, phosphate and sulfate. A summary of CEMRC QA/QC for inorganic analyses is presented in Appendix K. 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.

Gamma spectroscopy analysis was conducted using high purity Ge (HPGe) detector systems for 2-3 d. The systems have HPGe, p-type, coaxial detectors of ~50% nominal efficiency. A set of soil matrix standards was prepared using NIST traceable solutions and used

to establish matrix-specific calibration and counting efficiencies. For analyses of alpha-emitting radionuclides, 10-g aliquots of each sample were heated in a muffle furnace to combust organic material and spiked with a radioactive tracer to allow determination of the efficiency of extraction.

The samples for radiochemical analyses underwent total dissolution followed by NaOH fusion of the insoluble residues. Multiple precipitation, co-precipitation and ion-exchange and/or extraction chromatography procedures were then used to separate and purify the desired elements. The elements of interest were then precipitated with La, deposited onto filters, mounted and counted on an alpha spectroscopy system. A summary of CEMRC QA/QC for radioanalyses is presented in Appendix L.

Aliquots of 73 of the 96 samples collected in 1998 were analyzed by the NMSU SWAT laboratory for soil texture. Analyses were made using the hydrometer method (Soil Conservation Service, 1972, *Soil Survey Laboratory Methods and Procedures for Collecting Soil Samples*. SCS, USDA; Gee, G. W. and J. W. Bauder, 1986, *Particle-size Analysis*. In Kline, A. (ed.) *Methods of Soil Analysis. Part I. Physical and Mineralogical Methods - Agronomy Monograph no. 9*. American Society of Agronomy, Madison, WI) resulting in measurements of the percentages of sand, silt and clay.

Multivariate analysis of variance (MANOVA) was used to test for differences between the Cactus Flats and Near Field grids across analytes. Mean concentrations of all analytes were estimated by grid and by soil type and significant differences between means were identified using *t*-tests. Correlations of the concentrations of radionuclides to soil texture classes, and to concentrations of non-radioactive analytes, were computed using Pearson correlation coefficients. Student's *t*-tests were applied to pairwise differences between radionuclide concentrations reported by Duke Engineering (1998 CEMRC Report) and the concentrations determined by CEMRC.

## Results and Discussion

### **Baseline Analyses by CEMRC**

Results of the MANOVA showed that there were significant ( $p < 0.05$ ) differences between the two grids, and that the Cactus Flats grid generally had higher concentrations of metals than found on the Near Field grid. Of the inorganic analytes, 20 (Ag, Al, As, Ba, Be, Cd, Co, Cu, Eu, Fe, K, La, Mn, Ni, Pb, Th, Ti, U, V and Zn) showed significantly higher concentrations on the Cactus Flats grid as compared to the Near Field grid. Chloride showed a significantly lower concentration on the Cactus Flats grid.

Many metals, including radionuclides, are known to have an affinity for attaching to small particles in the soil (Muller, R. N. and D. G. Sprugel, 1977, *Health Physics* 33, 405; Muller, R. N. and G. T. Tissue, 1977, *Soil Science* 124, 191; Watters, R. L., et al., 1983, *Radiochemia Acta* 32, 89; Little, C. A., 1980, *Journal of Environmental Quality* 9, 350; Tamura, T., 1975, *J. Environ. Qual.* 4, 350). Clay minerals are aluminosilicates and hydrated oxides, and usually account for the major adsorptive component of soils (Wild, A., 1994, *Soils and the Environment*. Cambridge University Press; Whicker, F. W. and V. Schultz, 1982, *Radioecology: Nuclear Energy and the Environment. Vol. II*. CRC Press). Therefore, the concentration of aluminum in soil can be used as a surrogate for estimating the clay content of soils. A correlation between the concentration of Al and the percent clay in soils from 73 of the samples was highly significant ( $r = 0.51$ ,  $p < 0.001$ ), although the correlation between Al and silt was stronger ( $r = 0.69$ ,  $p < 0.001$ ). Similar correlations can be seen for many of the analytes (Table 12). Thus, comparing the ratios of the metals to Al helps to normalize for the effect of soil texture on the concentrations. The ratios of the metals to Al were similar between the two grids (Table 13). Only the ratios of Mo and Tl to Al were significantly ( $p < 0.01$ ) lower on the Cactus Flats grid.

The analyte:Al ratios in soils were compared to ratios reported herein for aerosols (Particulate Concentrations and Inorganics in Near Surface Air). Because aerosols are generally smaller than soil particles, the aerosols



tend to have higher mass concentrations of metals which adsorb to fine particles than do soils. The ratios of Ag, Ca, Cd, Co, Cr, Cu, La, Mn, Mo, Ni and V to Al were lower by a factor of 2 or more in the soils as compared to the aerosols at both Cactus Flats and Near Field. The ratios of Fe, Hg, K, Mg, Na and Th to Al were higher in the soils than in the aerosols by a factor of 2 or greater.

The average MDC for radionuclides determined by alpha spectroscopy was  $\approx 0.04$  mBq  $g^{-1}$ . The average MDC for radionuclides determined by gamma spectrometry was  $\approx 0.5$  mBq  $g^{-1}$ . Radionuclide activities greater than MDC were detected in all soil samples except for  $^{60}Co$ ,  $^{234m}Pa$  (1 value  $>MDC$ ),  $^{238}Pu$  (11 of 96  $>MDC$ ) and  $^{241}Am$  (72 of 96  $>MDC$ ). Only values greater than the MDC were included in the statistical analyses (Table 14). MANOVA confirmed that the Cactus Flats and Near Field grids were significantly ( $p < 0.05$ ) different. Concentrations of the radionuclides were significantly ( $p < 0.05$ ) higher on the Cactus Flats grid than on the Near Field grid for all radionuclides except  $^{238}Pu$  (Fig. 21). The mean  $^{239,240}Pu$  concentration ( $0.158$  mBq  $g^{-1}$ ) fell within the range reported by Kenny et al., (1995, *Radionuclide Baseline in Soil Near Project Gnome and the Waste Isolation Pilot Plant*, Environmental Evaluation Group, Carlsbad, New Mexico) at the WIPP ( $0 - 0.74$  mBq  $g^{-1}$ ) and was lower than background concentrations found at Hueston Woods and Urbana, Ohio ( $0.7 - 1.0$  mBq  $g^{-1}$ ) (Alberts J. J., et al., 1980, *J. Environ. Qual.* 9, 592) and at a series of 15 locations between Ft. Collins and Colorado Springs, Colorado ( $0.6 - 1.7$  mBq  $g^{-1}$ ) (Hodge, V. et al., 1996, *Chemosphere* 32, 2067).

The soil textures for all of the samples were very similar, with 81.2-95.8% sand, 1.5-13.4% silt and 1.4% to 5.7% clay. Eleven of the 73 samples were classified as loamy sand and the remainder as sand. All of the radionuclides except  $^{238}Pu$  showed significant ( $p < 0.01$ ) correlations with each of the soil texture classes (Table 12). In general, the percentage of sand, or equivalently, the sum of the percentages of silt and clay, was the best correlate for the concentration of radionuclides. However, for  $^{137}Cs$  and  $^{239,240}Pu$  concentrations the percent-

age of silt gave a slightly better correlation (Table 12).

In general, the mean ratios of concentrations of the radionuclides to Al were similar between the two grids. Only  $^{239,240}Pu$  showed a significant difference in these ratios, with the ratio for Cactus Flats being significantly greater ( $p < 0.05$ ) than that for Near Field. Concentrations of the radionuclides were also positively correlated by location with the concentrations of many of the analytes. All of the radionuclides were significantly ( $p < 0.01$ ) correlated with Al. Pb showed stronger correlations than Al for all radionuclides except  $^{235}U$ .

The correlation between  $^{40}K$  and  $^{39}K$  was only 0.625 (Fig. 22) and the average ratio ( $\pm 95\%$  CI) was 0.00165 ( $\pm 0.00009$ ) as compared to the expected isotopic ratio of 0.0117 (Browne, E. and R. B. Firestone, 1986, *Table of Radioactive Isotopes*, John Wiley & Sons, New York). The cause for this significant departure from the expected ratio and for the variability in the ratio is not known, but some of the variability may be due to the difference in the dissolution methods used for radioanalyses and inorganic analyses. The ratios of  $^{40}K$  to  $^{39}K$  are correlated with soil texture ( $r$  for percent sand = 0.369,  $r$  for percent silt = -0.325 and  $r$  for percent clay = -0.350;  $n = 96$ ,  $p < 0.01$ ). Because the half-life of  $^{40}K$  is about  $10^9$  years, it is unlikely that the effect on the ratios is due to input from decay-depleted sources, such as the local Permian-age potash deposits.

These results demonstrate that significant levels of variability in background levels of soil contaminants and constituents can occur in areas having relatively low variability in soil texture. The high correlation of the radionuclides and many of the non-radioactive metals to the percentages of sand, silt and clay in the soil explains much of the between-sample variability. Actinides can form strong complexes with oxygen-containing ligands (Beal, G. W. and B. Allard, 1981, In Tewari, P. H. (ed.), *Adsorption from Aqueous Solutions*, Plenum Press, New York; Allard, B., 1982, In Edlestein, N. M. (ed.), *Actinides in Perspective*, Pergamon Press, Oxford). The fact that  $^{239,240}Pu$  is more strongly correlated with the percentage of silt than with the percentage of clay suggests that many of the clay size parti-

cles provide less suitable binding sites for the adsorption of metals than those of typical clays. Actinides also form complexes with humic molecules and these can be significantly more stable than their complexes with simple inorganic ligands (Livens, F. R. and D. L. Singleton, 1991, *J. Environ. Rad.* 13, 323), although subject to a significant concentration effect (Hummel, W. et al., 1999, *Radiochim. Acta* 84, 111). Therefore, the affinity of <sup>239,240</sup>Pu with silt could also be due to a larger concentration of organic material in the silt than in the clay fractions. An investigation of soil particle mineralogy may help explain why the silt fraction is a better correlate of metal concentrations than is the clay fraction.

These data also suggest that the variability in concentrations across locations may arise from a redistribution of contaminated fine soil particles or from a greater degree of entrapment of the contaminants in the upper layer of the soil. Further elucidation of these complex relationships may be possible through selective soil profile analyses.

### **Inter- and Intra-Laboratory Comparisons**

Concentrations of two radioactive elements, uranium (<sup>234</sup>U, <sup>235</sup>U and <sup>238</sup>U) and thorium (<sup>232</sup>Th), were measured independently using ICP-MS and alpha counting methods. A comparison of the estimated concentrations for these analytes showed that the average ratio of the alpha estimate to the ICP-MS estimate for Th was 2.22. The average ratio of the alpha estimate of uranium to that of the ICP-MS method was 8.20. The larger estimates from the actinide methodology may reflect that the samples prepared for actinide analysis undergo more complete dissolution, including fusion of any residues, whereas the samples prepared for ICP-MS analysis undergo only acid leaching. In drinking water analyses reported herein (Radiological and Non-radiological Constituents in Selected Drinking Water Sources), where dissolved forms of the elements are expected to dominate, the average ratio ( $\pm$  SE) for U by the two methods was 1.02 ( $\pm$  0.09), but <sup>232</sup>Th was not detected in drinking water samples. A correlation analysis failed to show any relation between the alpha

counting to ICP-MS ratios and soil texture. If the difference in the estimates is due to the difference in dissolution, it appears that the refractory components are not strongly associated with a particular particle size class.

CEMRC measurements of radionuclides in soils were compared to previous measurements presented in the CEMRC 1997 Report performed by Accu-Labs Research, Inc. (Golden, Colorado) for samples collected during 1997 at the Near Field. Comparison of the means of the Accu-Lab and CEMRC results is somewhat difficult because of the limited precision of the Accu-Lab data. However, there were no significant ( $p < 0.05$ ) differences between the estimated activity concentrations except for <sup>230</sup>Th, for which the difference between the Accu-Lab mean (14.8 mBq g<sup>-1</sup>) and the CEMRC mean (10.4 mBq g<sup>-1</sup>) was significant at  $p < 0.001$ . The mean <sup>239,240</sup>Pu activity concentration ( $\pm$  95% CI) of the 48 samples measured by CEMRC for the Near Field grid was 0.101 ( $\pm$  0.014) mBq g<sup>-1</sup>. This mean is not significantly different from the mean of the Accu-Lab 1997 results [0.14 ( $\pm$  0.041) mBq g<sup>-1</sup>].

CEMRC measurements of radionuclides in soils were also compared to measurements presented in the CEMRC 1998 Report performed by DES. Comparisons used (1) a paired t-test on the difference of the CEMRC and DES results paired by sample and (2) correlations between the DES and CEMRC results. The paired t-test showed a significant difference only between the estimates of <sup>232</sup>Th concentrations ( $p < 0.05$ ,  $n = 36$ ) (Table 15). The DES results for each radionuclide were found to be uncorrelated with the Accu-Lab results when paired by location (CEMRC 1998 Report) but were well correlated with the CEMRC results when paired by sample. It should be noted that the DES/CEMRC comparison is based on pairing aliquots from the same samples, not on independent samples from the same grid locations, as was the case with the Accu-Labs/DES comparison.

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

**Table 12. Correlations between Analytes and Soil Texture Components**

Analyte	<sup>a,b</sup> Correlation Coefficients			Analyte	Correlation Coefficients		
	% Sand	% Silt	% Clay		% Sand	% Silt	% Clay
<sup>137</sup> Cs	-.87	.88	.49	Chloride	<b>.19</b>	<b>-.19</b>	<b>-.11</b>
<sup>208</sup> Tl	-.91	.89	.63	Co	-.77	.69	.70
<sup>212</sup> Bi	-.88	.83	.66	Cr	-.37	.29	.45
<sup>212</sup> Pb	-.90	.86	.66	Cu	-.80	.74	.67
<sup>214</sup> Bi	-.85	.83	.59	Eu	-.66	.59	.62
<sup>214</sup> Pb	-.87	.85	.59	Fe	-.73	.68	.57
<sup>228</sup> Ac	-.92	.89	.64	K	-.73	.71	.48
<sup>228</sup> Th	-.93	.90	.66	La	-.71	.63	.65
<sup>230</sup> Th	-.90	.88	.60	Li	-.39	.32	.43
<sup>232</sup> Th	-.93	.89	.67	Mg	-.58	.56	.39
<sup>234</sup> U	-.74	.71	.54	Mn	-.74	.72	.52
<sup>235</sup> U	-.49	.51	.24	Ni	-.69	.63	.60
<sup>238</sup> U	-.74	.70	.54	Pb	-.81	.73	.69
<sup>239,240</sup> Pu	-.84	.85	.49	Sb	<b>.02</b>	<b>-.07</b>	<b>.11</b>
<sup>40</sup> K	-.78	.77	.48	Sr	<b>-.23</b>	.18	.29
Ag	-.28	.28	<b>.16</b>	Th	-.71	.63	.68
Al	-.71	.69	.51	Ti	-.49	.43	.48
As	-.67	.59	.63	U	-.58	.49	.63
Ba	-.49	.43	.45	V	-.45	.32	.63
Ca	-.25	.25	<b>.16</b>	Zn	-.73	.68	.57
Cd	-.63	.59	.48				

<sup>a</sup>Pearson product moment correlation coefficients; all correlations are significant at p<0.05 except those in shaded cells

<sup>b</sup>Correlations for radionuclides based on 73 samples; correlations for other constituents based on 66 samples

**Table 13. Mean Ratios of Analytes to Al in Soil Samples  
from Cactus Flats and Near Field Grids**

Analyte	Near Field	<sup>a</sup> N	Cactus Flats	N	Analyte	Near Field	N	Cactus Flats	N
<sup>137</sup> Cs	2.1E-03	48	3.0E-03	48	Cu	1.2E-03	48	1.2E-03	48
<sup>208</sup> Tl	1.9E-03	48	2.0E-03	48	Eu	5.4E-05	48	5.2E-05	48
<sup>212</sup> Bi	6.0E-03	48	6.7E-03	48	Fe	1.5E+00	48	1.6E+00	48
<sup>212</sup> Pb	5.6E-03	48	5.9E-03	48	Hg	2.9E-03	22	3.9E-03	13
<sup>214</sup> Bi	5.7E-03	48	5.7E-03	48	K	2.9E-01	48	3.3E-01	48
<sup>214</sup> Pb	5.9E-03	48	6.0E-03	48	La	1.8E-03	48	1.9E-03	48
<sup>228</sup> Ac	5.6E-03	48	6.0E-03	48	Li	1.2E-03	48	1.0E-03	48
<sup>228</sup> Th	5.9E-03	48	6.2E-03	48	Mg	2.1E-01	48	2.1E-01	48
<sup>230</sup> Th	6.2E-03	48	6.4E-03	48	Mn	2.1E-02	48	2.4E-02	48
<sup>232</sup> Th	5.6E-03	48	6.0E-03	48	Mo	8.7E-05	29	5.1E-05	38
<sup>234</sup> U	5.2E-03	48	4.8E-03	48	Na	3.6E-02	7	8.2E-02	9
<sup>235</sup> U	2.7E-04	48	2.6E-04	48	Ni	1.1E-03	48	1.2E-03	48
<sup>238</sup> Pu	3.3E-05	4	2.0E-05	7	Nitrate	5.4E-03	48	5.7E-03	48
<sup>238</sup> U	5.4E-03	48	5.0E-03	48	Nitrite	8.9E-05	4	2.0E-04	5
<sup>239,240</sup> Pu	6.9E-05	48	1.1E-04	48	Pb	1.6E-03	48	1.9E-03	48
<sup>241</sup> Am	3.5E-05	31	4.3E-05	40	Phosphate	3.3E-03	36	3.5E-03	41
<sup>40</sup> K	1.4E-01	48	1.3E-01	48	Sb	4.8E-05	46	3.3E-05	48
Ag	8.5E-06	46	1.0E-05	48	Se	4.2E-05	3	3.9E-05	5
As	4.9E-04	48	6.1E-04	48	Sr	3.1E-03	48	2.6E-03	48
Ba	1.3E-02	48	1.4E-02	48	Sulfate	5.8E-03	48	4.8E-03	48
Be	6.9E-05	48	7.6E-05	48	Th	6.4E-04	48	6.8E-04	48
Ca	3.1E-01	48	3.0E-01	45	Ti	3.4E-02	48	3.8E-02	48
Cd	3.8E-05	43	4.0E-05	48	Tl	2.7E-04	15	1.4E-04	10
Chloride	2.3E-03	48	1.0E-03	48	U	5.3E-05	48	5.1E-05	48
Co	3.8E-04	48	4.0E-04	48	V	2.3E-03	48	2.2E-03	48
Cr	2.0E-03	47	1.8E-03	48	Zn	5.2E-03	48	6.4E-03	48

<sup>a</sup>N = number of samples

**Table 14. Summary Statistics for Analytes in Soil Samples Collected in 1998**

Analyte	Unit	Cactus Flats				Near Field			
		<sup>a</sup> N	<sup>b</sup> Mean	<sup>c</sup> SE	Range	N	Mean	SE	Range
<sup>137</sup> Cs	mBq g <sup>-1</sup>	48	6.2E+00	5.2E-01	6.9E-01 - 1.5E+01	48	3.1E+00	2.3E-01	3.1E-01 - 6.0E+00
<sup>208</sup> Tl	mBq g <sup>-1</sup>	48	3.6E+00	1.1E-01	2.4E+00 - 5.4E+00	48	2.7E+00	7.8E-02	1.7E+00 - 3.7E+00
<sup>212</sup> Bi	mBq g <sup>-1</sup>	48	1.2E+01	3.7E-01	6.5E+00 - 1.9E+01	48	8.7E+00	2.8E-01	5.2E+00 - 1.5E+01
<sup>212</sup> Pb	mBq g <sup>-1</sup>	48	1.1E+01	3.2E-01	7.3E+00 - 1.7E+01	48	8.3E+00	2.5E-01	5.4E+00 - 1.1E+01
<sup>214</sup> Bi	mBq g <sup>-1</sup>	48	1.0E+01	2.8E-01	7.1E+00 - 1.5E+01	48	8.4E+00	2.2E-01	5.7E+00 - 1.1E+01
<sup>214</sup> Pb	mBq g <sup>-1</sup>	48	1.1E+01	2.9E-01	7.8E+00 - 1.6E+01	48	8.7E+00	2.2E-01	6.1E+00 - 1.1E+01
<sup>228</sup> Ac	mBq g <sup>-1</sup>	48	1.1E+01	3.4E-01	6.7E+00 - 1.6E+01	48	8.2E+00	2.4E-01	5.6E+00 - 1.1E+01
<sup>228</sup> Th	mBq g <sup>-1</sup>	48	1.1E+01	3.5E-01	7.1E+00 - 1.8E+01	48	8.7E+00	2.3E-01	6.0E+00 - 1.2E+01
<sup>230</sup> Th	mBq g <sup>-1</sup>	48	1.2E+01	3.9E-01	7.7E+00 - 2.0E+01	48	9.1E+00	2.4E-01	6.3E+00 - 1.3E+01
<sup>232</sup> Th	mBq g <sup>-1</sup>	48	1.1E+01	3.2E-01	6.6E+00 - 1.6E+01	48	8.2E+00	2.2E-01	5.7E+00 - 1.1E+01
<sup>234</sup> U	mBq g <sup>-1</sup>	48	8.6E+00	2.8E-01	4.9E+00 - 1.2E+01	48	7.5E+00	2.0E-01	5.0E+00 - 1.0E+01
<sup>235</sup> U	mBq g <sup>-1</sup>	48	4.8E-01	4.3E-02	2.3E-01 - 2.4E+00	48	3.9E-01	1.3E-02	2.1E-01 - 5.7E-01
<sup>238</sup> Pu	mBq g <sup>-1</sup>	7	2.2E-02	3.3E-03	1.1E-02 - 3.2E-02	4	4.5E-02	1.7E-02	1.6E-02 - 8.5E-02
<sup>238</sup> U	mBq g <sup>-1</sup>	48	8.9E+00	3.0E-01	4.9E+00 - 1.3E+01	48	7.7E+00	2.2E-01	5.3E+00 - 1.1E+01
<sup>239,240</sup> Pu	mBq g <sup>-1</sup>	48	2.2E-01	1.8E-02	2.7E-02 - 5.1E-01	48	1.0E-01	6.9E-03	1.5E-02 - 1.9E-01
<sup>241</sup> Am	mBq g <sup>-1</sup>	40	8.3E-02	6.9E-03	2.1E-02 - 2.6E-01	31	4.9E-02	4.4E-03	1.7E-02 - 1.3E-01
<sup>40</sup> K	mBq g <sup>-1</sup>	48	2.3E+02	6.4E+00	1.4E+02 - 3.2E+02	48	2.1E+02	5.5E+00	1.4E+02 - 2.8E+02
Ag	mg kg <sup>-1</sup>	48	1.8E-02	1.5E-03	6.5E-03 - 7.6E-02	46	1.3E-02	1.1E-03	5.2E-03 - 4.1E-02
Al	mg kg <sup>-1</sup>	48	2.4E+03	1.5E+02	1.5E+02 - 6.0E+03	48	1.7E+03	9.5E+01	7.9E+02 - 3.6E+03
As	mg kg <sup>-1</sup>	48	1.2E+00	5.0E-02	5.1E-01 - 2.3E+00	48	7.4E-01	2.6E-02	4.6E-01 - 1.3E+00
Ba	mg kg <sup>-1</sup>	48	2.5E+01	1.8E+00	7.6E+00 - 8.5E+01	48	1.9E+01	2.0E+00	5.4E+00 - 8.5E+01
Be	mg kg <sup>-1</sup>	48	1.5E-01	6.5E-03	6.9E-02 - 2.7E-01	48	1.0E-01	5.0E-03	4.9E-02 - 1.8E-01
Ca	mg kg <sup>-1</sup>	45	6.3E+02	5.4E+01	2.6E+02 - 2.0E+03	48	5.3E+02	8.0E+01	2.4E+02 - 3.7E+03
Cd	mg kg <sup>-1</sup>	48	7.9E-02	5.4E-03	3.1E-02 - 1.9E-01	43	5.9E-02	5.3E-03	2.7E-02 - 1.9E-01
Chloride	mg kg <sup>-1</sup>	48	1.8E+00	1.8E-01	7.5E-01 - 9.2E+00	48	3.1E+00	2.2E-01	7.0E-01 - 8.4E+00
Co	mg kg <sup>-1</sup>	48	8.0E-01	4.0E-02	3.0E-01 - 1.5E+00	48	5.8E-01	3.0E-02	2.3E-01 - 1.1E+00
Cr	mg kg <sup>-1</sup>	48	3.4E+00	1.4E-01	1.3E+00 - 6.0E+00	47	3.0E+00	1.9E-01	1.3E+00 - 9.1E+00
Cu	mg kg <sup>-1</sup>	48	2.3E+00	9.1E-02	1.3E+00 - 3.8E+00	48	1.8E+00	1.1E-01	7.7E-01 - 5.7E+00
Eu	mg kg <sup>-1</sup>	48	1.0E-01	5.1E-03	4.1E-02 - 2.1E-01	48	8.1E-02	4.7E-03	4.0E-02 - 2.1E-01
Fe	mg kg <sup>-1</sup>	48	3.1E+03	1.3E+02	1.7E+03 - 5.6E+03	48	2.3E+03	1.1E+02	1.2E+03 - 4.1E+03
Hg	mg kg <sup>-1</sup>	13	7.1E+00	2.1E+00	2.6E+00 - 3.1E+01	22	4.3E+00	4.7E-01	2.5E+00 - 1.1E+01
K	mg kg <sup>-1</sup>	48	6.6E+02	4.1E+01	3.2E+02 - 1.6E+03	48	4.7E+02	2.6E+01	2.4E+02 - 8.9E+02
La	mg kg <sup>-1</sup>	48	3.6E+00	1.5E-01	1.6E+00 - 6.5E+00	48	2.7E+00	1.4E-01	1.4E+00 - 6.5E+00
Li	mg kg <sup>-1</sup>	48	2.0E+00	8.8E-02	9.2E-01 - 3.3E+00	48	1.7E+00	1.4E-01	6.2E-01 - 4.5E+00
Mg	mg kg <sup>-1</sup>	48	4.4E+02	3.3E+01	1.8E+02 - 1.2E+03	48	3.6E+02	2.8E+01	1.5E+02 - 1.2E+03
Mn	mg kg <sup>-1</sup>	48	4.6E+01	2.9E+00	2.2E+01 - 1.3E+02	48	3.2E+01	1.7E+00	1.4E+01 - 6.3E+01
Mo	mg kg <sup>-1</sup>	38	1.2E-01	4.6E-03	7.6E-02 - 1.8E-01	29	1.3E-01	1.4E-02	7.7E-02 - 4.7E-01
Na	mg kg <sup>-1</sup>	9	7.0E+01	7.4E+00	3.7E+01 - 1.1E+02	7	6.7E+01	3.3E+00	5.8E+01 - 7.6E+01
Ni	mg kg <sup>-1</sup>	48	2.3E+00	1.2E-01	9.8E-01 - 5.4E+00	48	1.6E+00	9.2E-02	6.2E-01 - 3.8E+00
Nitrate	mg kg <sup>-1</sup>	48	8.2E+00	7.0E-01	3.2E+00 - 2.8E+01	48	7.6E+00	6.0E-01	2.4E+00 - 2.1E+01

Table continued on next page

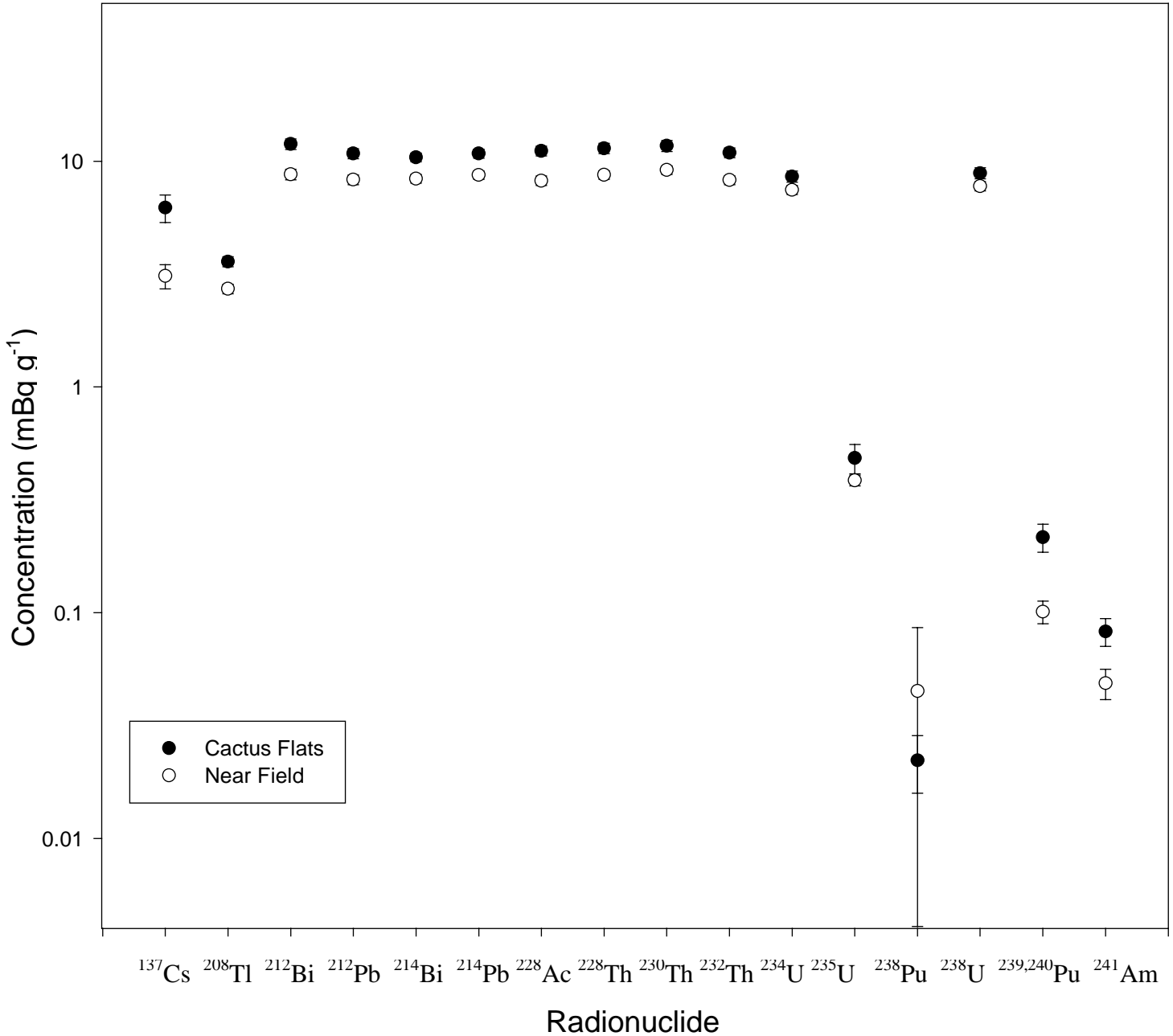
**Table 14. Summary Statistics for Analytes in Soil Samples Collected in 1998  
(Continued)**

Analyte	Unit	Cactus Flats				Near Field			
		<sup>a</sup> N	<sup>b</sup> Mean	<sup>c</sup> SE	Range	N	Mean	SE	Range
Nitrite	mg kg <sup>-1</sup>	5	1.5E-01	2.2E-02	9.0E-02 - 2.2E-01	4	1.0E-01	9.5E-03	9.0E-02 - 1.3E-01
Pb	mg kg <sup>-1</sup>	48	3.5E+00	1.5E-01	1.5E+00 - 5.9E+00	48	2.4E+00	1.1E-01	1.3E+00 - 4.2E+00
Phosphate	mg kg <sup>-1</sup>	41	5.5E+00	3.5E-01	1.3E+00 - 1.0E+01	36	4.8E+00	3.0E-01	2.5E+00 - 1.0E+01
Sb	mg kg <sup>-1</sup>	48	6.1E-02	3.3E-03	2.2E-02 - 1.1E-01	46	6.7E-02	8.8E-03	2.6E-02 - 2.6E-01
Se	mg kg <sup>-1</sup>	5	1.2E-01	1.2E-02	1.0E-01 - 1.6E-01	3	1.1E-01	2.7E-02	5.6E-02 - 1.4E-01
Sr	mg kg <sup>-1</sup>	48	4.9E+00	1.0E+00	1.7E+00 - 5.2E+01	48	4.8E+00	1.1E+00	1.4E+00 - 5.2E+01
Sulfate	mg kg <sup>-1</sup>	48	8.7E+00	5.7E-01	2.8E+00 - 2.1E+01	48	8.1E+00	4.3E-01	2.9E+00 - 1.6E+01
Th	mg kg <sup>-1</sup>	48	1.3E+00	5.3E-02	5.5E-01 - 2.1E+00	48	9.6E-01	4.4E-02	4.8E-01 - 1.5E+00
Ti	mg kg <sup>-1</sup>	48	6.6E+01	2.6E+00	2.5E+01 - 1.2E+02	48	5.1E+01	3.4E+00	2.2E+01 - 1.2E+02
Tl	mg kg <sup>-1</sup>	10	2.7E-01	5.0E-02	1.2E-01 - 5.7E-01	15	3.5E-01	4.2E-02	1.2E-01 - 5.9E-01
U	mg kg <sup>-1</sup>	48	9.6E-02	4.4E-03	4.0E-02 - 1.6E-01	48	8.0E-02	3.6E-03	3.9E-02 - 1.6E-01
V	mg kg <sup>-1</sup>	48	4.2E+00	1.7E-01	2.1E+00 - 6.5E+00	48	3.5E+00	1.3E-01	1.6E+00 - 6.0E+00
Zn	mg kg <sup>-1</sup>	48	1.2E+01	6.0E-01	5.7E+00 - 2.4E+01	48	7.8E+00	4.3E-01	2.9E+00 - 1.5E+01

<sup>a</sup>N = number of samples<sup>b</sup>Mean = arithmetic mean<sup>c</sup>SE = standard error of mean**Table 15. Differences Between CEMRC and Duke Engineering Measurements  
of Radionuclides in Soil Samples Collected in 1998**

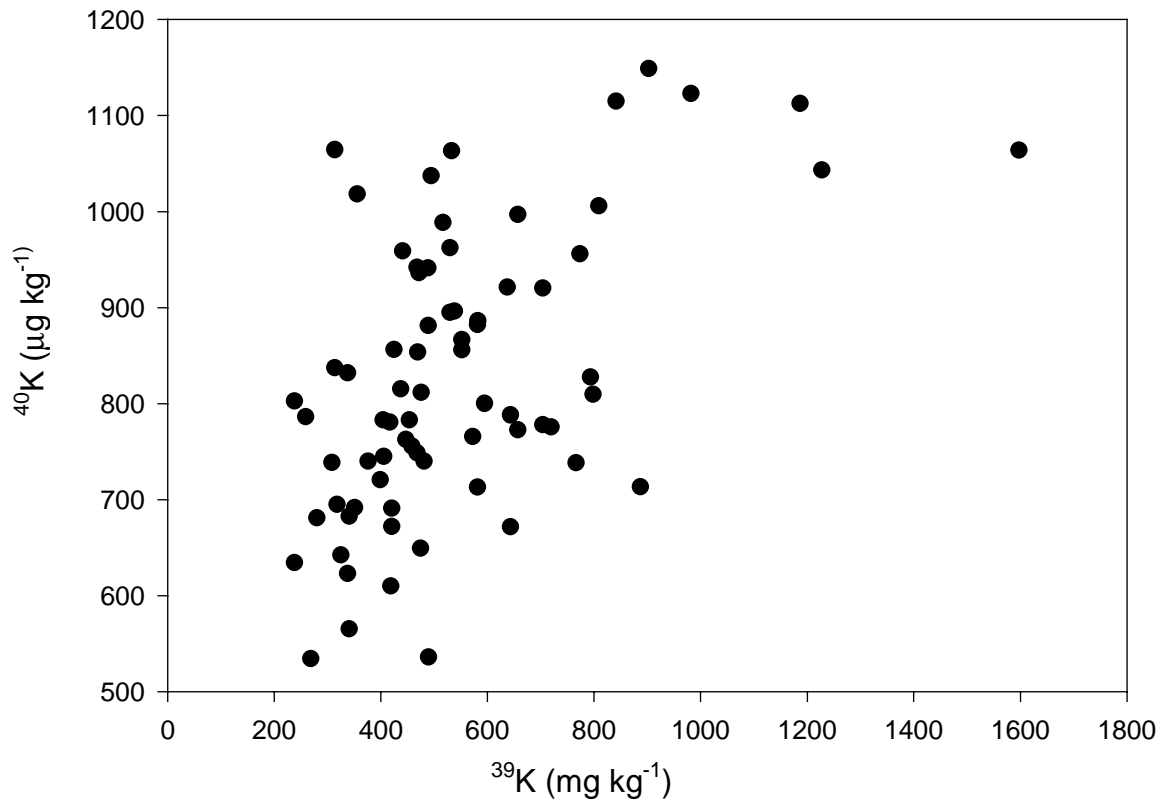
Analyte	<sup>a</sup> N	<sup>b</sup> Mean of Paired Differences (mBq g <sup>-1</sup> )	<sup>c</sup> SE
<sup>239,240</sup> Pu	37	1.0E-02	7.5E-03
<sup>238</sup> U	37	-2.5E-01	3.0E-01
<sup>235</sup> U	37	7.6E-03	1.9E-02
<sup>234</sup> U	37	-3.2E-01	2.7E-01
<sup>232</sup> Th	37	-7.0E-01	2.8E-01
<sup>230</sup> Th	37	3.8E-01	3.6E-01
<sup>228</sup> Th	37	-1.0E-01	3.2E-01

<sup>a</sup>N = number of comparisons included in calculation<sup>b</sup>Mean difference = value for analyte measured by CEMRC minus value for analyte measured by Duke Engineering, summed across N split samples, divided by N<sup>c</sup>SE = standard error of mean



**Figure 21. Mean Radionuclide Activity Concentration in Soil Samples from Near Field and Cactus Flats Grids**

Error bars are 95% confidence intervals of the mean.



**Figure 22. Concentrations of  $^{40}\text{K}$  versus  $^{39}\text{K}$  in Soil Samples from the Near Field and Cactus Flats Grids**



## Radiological and Non-radiological Constituents in Surface Water and Sediments at Selected Reservoirs

### Introduction

As part of the WIPP EM programs, surface water and sediments are routinely sampled from three regional reservoirs including Brantley Lake, Lake Carlsbad and Red Bluff Reservoir. Brantley Lake and Red Bluff Reservoir were selected for sampling because they are impoundments located “upstream” and “downstream”, respectively, relative to surface and ground water flows from the area immediately surrounding the WIPP site. Both reservoirs support a warm-water fishery and are used for irrigation, livestock watering, wildlife habitat and recreation. Lake Carlsbad is an impounded section of the Pecos River within the city of Carlsbad that is used extensively by the local population for recreational warm-water fishing, boating and swimming. In addition, it can be used for industrial water supply, livestock watering and wildlife habitat (20 NMAC 6.1, 1995, State of New Mexico Standards for Interstate and Intrastate Streams).

In 1997, a pilot study of the surface water and sediments in Brantley Lake was conducted, in which 15 sediment and three surface water samples were collected during March and April and three additional water samples were collected in September. A summary of the sample analyses was included in the 1997 CEMRC Annual Report. In 1998, 24 sediment and 17 surface water samples were collected from Brantley Lake, Lake Carlsbad and Red Bluff Reservoir. These included 12 sediment samples and 11 surface water samples collected during January - April and the remaining samples (12 sediment and 6 surface water) collected during August - October. The results of actinide, elemental, inorganic and selected organic analyses of the first set of samples collected in 1998 were reported in the 1998 CEMRC Report. In 1999, six surface water and 12 sediment samples were collected from the three reservoirs during June - July.

Analyses reported herein summarize the baseline results for radiological constituents in

regional sediment and surface water and results from analyses of gamma-emitting radionuclides from the first monitoring phase samples collected in 1999. The baseline inorganic analyses summary is updated to include the latest surface water and sediment measurements.

### Methods

Sediment samples are routinely collected at randomly selected locations within the deep basins of each reservoir (Fig. 23, 24 and 25). Deep basins were chosen for sampling to minimize the disturbance and particle mixing effects of current and wave action that occur at shallower depths. Also, many of the analytes of interest tend to concentrate in the fine sediments that settle in the deep reservoir basins; thus, measurements from these areas would typically represent the highest levels that might be expected for a given reservoir.

Sediments were collected to depths of 5-10 cm using a grab sampler or Eckman dredge, to obtain  $\geq 6$  L of sediment at each sampling site. In the field,  $\geq 5$  L of each sample was sealed in a pre-cleaned plastic bucket and transported to CEMRC for preparation prior to analyses.

In the laboratory, the sediment samples were homogenized and split into aliquots for various analyses. Samples destined for radiochemical analyses were dried at 105°C to a constant weight, pulverized and homogenized prior to analysis. Samples analyzed for elemental and inorganic constituents were dried at 60 °C, and pulverized prior to analysis. A 250-g aliquot of each sample collected in 1998 was sent to Duke Engineering & Services (Bolton, Massachusetts) where it was analyzed for the alpha-emitting radionuclides  $^{239,240}\text{Pu}$ ,  $^{228}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$ . Inorganic analyses were produced by CEMRC using IC, ICP-ES, ICP-MS and AAS, with methods described elsewhere in this report.

In addition to the above analyses, 1-L aliquots of the 12 sediment samples collected in 1999 were sent to the NMSU SWAT laboratory for particle size and texture analysis (Soil

Conservation Service, 1972. *Soil Survey Laboratory Methods and Procedures for Collecting Soil Samples*. SCS, USDA; Gee, G. W. and J. W. Bauder, 1986. *Particle-size Analysis*. In Kline, A. (ed.) *Methods of Soil Analysis. Part I. Physical and Mineralogical Methods - Agronomy Monograph no. 9*. American Society of Agronomy, Madison, WI).

For 1999 studies, surface water was collected at one location within each reservoir. The surface water samples were taken in the same general area as the sediment samples. At each sampling location, one sample was collected from the surface (~ 0.5 - 1 m depth) and a second sample was collected from approximately 0.5 - 1 m above the sediment bed. The sample volume collected for radiological analyses was approximately 5 L. In addition, two 1-L samples were collected for inorganic analyses.

In the laboratory, surface water samples collected for radiological analyses were vacuum-filtered to 0.2  $\mu\text{m}$ , acidified with  $\text{HNO}_3$  to a  $\text{pH} < 2$ , and a 3-L aliquot removed for analysis of actinide and gamma-emitting radionuclides. Samples collected for elemental analyses were prepared according to the applicable EPA standard methods for the instrumentation used.

## Results

### **Baseline Radiological Analyses of Deep Basin Reservoir Sediments**

Baseline actinide analyses ( $^{239,240}\text{Pu}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{228}\text{Th}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$ ) have been completed on 29 deep basin sediment samples collected through the end of 1998 including 13 sediment samples from Brantley Lake, eight samples from Lake Carlsbad and eight samples from Red Bluff Reservoir. Across all three reservoirs, baseline  $^{239,240}\text{Pu}$  activity concentrations in individual sediment samples ranged from 0.07 to 0.52  $\text{mBq g}^{-1}$  with a regional mean ( $\pm\text{SE}$ ) concentration of 0.26 ( $\pm 0.02$ )  $\text{mBq g}^{-1}$ . ANOVA showed there were significant differences ( $p < 0.05$ ) in the mean  $^{239,240}\text{Pu}$  activity concentrations between the individual reservoirs (Fig. 26 and Table 16) with the lowest mean concentration in Lake Carlsbad and highest mean concentration in

Red Bluff reservoir. Within reservoirs, the mean  $^{239,240}\text{Pu}$  activity concentration has remained relatively stable over the baseline sampling interval with none of the reservoirs exhibiting an obvious increasing or decreasing trend over time.

Sediment activity concentrations of  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$  ranged from 9.9 to 123.0  $\text{mBq g}^{-1}$ , 1.2 to 3.7  $\text{mBq g}^{-1}$  and 7.5 to 71.7  $\text{mBq g}^{-1}$ , respectively. Mean sediment activity concentrations for all three U isotopes were similar between Lake Carlsbad and Brantley Lake but were almost a factor of 2 higher in Red Bluff Reservoir (Fig. 27). (U concentrations in sediment samples measured by ICP-MS were also approximately twice as high at Red Bluff Reservoir as at the other two lakes.) U activity concentrations in sediment across all reservoirs have exhibited a slight increase over time in all reservoirs, but the number of data points is too small to document a significant trend.

Across all reservoirs, Th sediment activity concentrations in individual samples ranged from 11.1 to 82.1  $\text{mBq g}^{-1}$ , 14.4 to 96.9  $\text{mBq g}^{-1}$  and 11.9 to 96.0  $\text{mBq g}^{-1}$  for  $^{228}\text{Th}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$ , respectively. However, the mean activity concentration for all of the Th isotopes was relatively similar across the baseline sampling period in all three reservoirs (Fig. 28).

Although the sediment concentrations of the U and Th isotopes were variable within and between reservoirs, the isotopic ratios were very similar across all three reservoirs. All three reservoirs appeared to be slightly enriched in  $^{234}\text{U}$  compared to  $^{238}\text{U}$ , with mean  $^{234}\text{U}/^{238}\text{U}$  activity ratios ranging from 1.4 to 1.6. Mean  $^{228}\text{Th}/^{232}\text{Th}$  ratios were close to unity, ranging from 0.75 to 1.1, indicating that these isotopes are in secular equilibrium within the sediments. A discussion of this phenomenon and the behavior of U and Th in freshwater systems is included in the CEMRC 1998 Report.

### **Baseline Radiological Analyses of Surface Waters**

Filtered surface water samples collected during 1998 were analyzed by gamma spectroscopy for 11 naturally occurring and 12 anthropogenic gamma-emitting radionuclides. With the exception of two Red Bluff Reservoir

samples that had detectable levels of  $^{40}\text{K}$ , all results were below detection limits. However, radioactivity may be present in the water column in association with the seston (suspended particulate or colloids larger than  $0.2\ \mu\text{m}$ ) that was filtered out and not included in the analyses.

A 200-L sample of unfiltered surface water was collected in Brantley Lake and Red Bluff Reservoir in 1998 and sent to Los Alamos National Laboratory for  $^{239}\text{Pu}$  analysis using thermal ionization mass spectroscopy (TIMS).  $^{239}\text{Pu}$  was not detected at MDC's of  $1.3\ \mu\text{Bq L}^{-1}$  for the Brantley Lake sample and  $2.2\ \mu\text{Bq L}^{-1}$  for the Red Bluff Reservoir sample.

### ***Monitoring Phase Radiological Analyses of Surface Water***

In 1999, two surface water samples (surface and deep positions) were collected from a single location in each reservoir. The samples were filtered in the laboratory and analyzed by gamma spectroscopy for gamma-emitting radionuclides. All results were below detection limits for these analytes. Actinide analyses of the samples are still pending and results will be reported, as they become available.

### ***Baseline Non-Radiological Analyses of Surface Water and Sediment***

To date, 16 surface water samples have been analyzed for a suite of inorganic compounds by CEMRC, including six samples collected in 1999. The majority of the analytes were detected in at least one sample collected in all three reservoirs (Table 17). However, five analytes, Cd, phosphate, Ag, Sn and Tl, have not been measured above detection limits in any of the samples collected to date. Be has been detected in two samples collected in Red Bluff but has not been detected in any of the Lake Carlsbad or Brantley Lake samples. Pb and nitrates have been detected in at least one Lake Carlsbad sample but have not

been above detection limits in any of the Brantley Lake or Red Bluff Reservoir samples. Hg has not been detected in any Lake Carlsbad samples to date, but has been above detection limits in 3 out of 5 Red Bluff and 4 out of 5 Brantley Lake samples.

To date, 36 sediment samples (12 from each reservoir) have been analyzed by CEMRC for the same suite of inorganic constituents as surface water samples. Most of the analytes were detected in all of the sediment samples from the three reservoirs (Table 18). However, fluoride and Sn have been below the detection limits in all of the sediment samples analyzed to date. Nitrate has been detected in one sample collected in Lake Carlsbad but not in any of the Red Bluff Reservoir or Brantley Lake sediments. Tl was detected in approximately half of the samples collected and analyzed in Brantley Lake and Lake Carlsbad but has not been above detection limits in any of the Red Bluff Reservoir samples analyzed to date. Phosphate has been detected in 5 out of 12 Lake Carlsbad samples but has not been detected in any of the Brantley Lake or Red Bluff Reservoir samples.

Texture analyses have been completed on five samples collected from each reservoir. The particle size distribution of the sediment samples is similar between all the reservoirs, with small particles dominating the sediment composition. Silt and clay size particles combined (sum of the mean percentages) made up 87%, 78% and 77% of Brantley Lake, Lake Carlsbad and Red Bluff sediment samples, respectively.

Tables presenting the surface water and sediment data summarized herein are available on the CEMRC web site at <http://www.cemrc.org>.

**Table 16. Summary Statistics for Selected Actinide Activity Concentrations in Sediment Samples Collected from Three Regional Reservoirs during 1997-1998**

Radionuclide	Activity Concentration (mBq g <sup>-1</sup> )											
	Brantley Lake				Red Bluff Reservoir				Lake Carlsbad			
	<sup>a</sup> N	<sup>b</sup> Mean	<sup>c</sup> SE	<sup>d</sup> CV (%)	N	Mean	SE	CV (%)	N	Mean	SE	CV (%)
<sup>239,240</sup> Pu	<sup>e</sup> 12	0.26	0.01	17	8	0.37	0.04	28	<sup>f</sup> 7	0.15	0.02	35
<sup>228</sup> Th	13	36.05	4.58	46	8	32.94	1.05	9	8	23.35	2.82	34
<sup>230</sup> Th	13	42.61	5.81	49	8	34.17	1.35	11	8	25.25	1.77	20
<sup>232</sup> Th	13	39.60	6.29	57	8	28.72	0.88	9	8	31.18	8.73	79
<sup>234</sup> U	13	43.94	5.47	45	8	91.04	8.35	26	8	43.66	2.42	16
<sup>235</sup> U	<sup>g</sup> 8	1.99	0.11	15	8	2.88	0.24	24	8	1.51	0.06	11
<sup>238</sup> U	13	32.22	3.45	39	8	55.36	4.45	23	8	29.49	1.16	11

<sup>a</sup>N = number of samples ; only values > MDC included in calculations

<sup>b</sup>Mean = arithmetic mean

<sup>c</sup>SE = standard error of mean

<sup>d</sup>CV = coefficient of variation; standard deviation expressed as percentage of the mean; CVs may reflect small rounding error

<sup>e</sup>Analysis failed on one sample collected on 3/20/97

<sup>f</sup>Analysis failed on one sample collected on 8/19/98

<sup>g</sup><sup>235</sup>U measurements were not made on sediment samples collected in 1997 at Brantley Lake

**Table 17. Summary Statistics for Elemental Constituents in Surface Water Samples Collected in 1999 from Three Regional Reservoirs**

Analyte	Concentration (mg L <sup>-1</sup> )								
	Brantley Lake			Red Bluff Reservoir			Lake Carlsbad		
	<sup>a</sup> N	<sup>b</sup> Mean	<sup>c</sup> SE	N	Mean	SE	N	Mean	SE
Ag	0	<sup>d</sup> NA	NA	0	NA	NA	0	NA	NA
Al	3	7.18E-02	3.51E-02	2	6.81E-02	4.90E-02	4	2.32E-01	6.43E-02
As	5	3.31E-03	1.28E-03	5	3.26E-03	6.82E-04	6	1.76E-03	1.37E-04
Ba	5	4.63E-02	1.12E-02	5	6.25E-02	5.24E-03	6	2.83E-02	4.78E-03
Be	0	NA	NA	2	3.56E-05	2.65E-06	0	NA	NA
Ca	5	2.86E+02	7.37E+01	5	3.78E+02	3.91E+01	6	3.11E+02	5.23E+00
Cd	0	NA	NA	0	NA	NA	0	NA	NA
Ce	3	1.28E-04	7.05E-05	2	5.17E-05	1.24E-05	2	2.48E-04	1.68E-04
Chloride	4	1.22E+03	4.14E+02	5	1.66E+03	1.78E+02	6	6.13E+02	1.02E+02
Co	5	3.86E-03	2.61E-03	5	4.20E-03	2.65E-03	4	6.83E-03	2.66E-03
Cr	2	7.44E-04	1.17E-04	4	1.28E-03	6.10E-04	2	1.00E-03	3.89E-04
Cu	3	7.19E-03	2.07E-03	5	6.37E-03	1.11E-03	4	4.30E-03	1.23E-03
Dy	2	1.46E-05	8.78E-06	2	4.07E-06	6.00E-08	2	2.09E-05	1.42E-05
Er	2	8.02E-06	4.50E-06	2	2.66E-06	5.75E-07	1	1.51E-05	NA
Eu	4	1.31E-05	3.86E-06	4	2.12E-05	2.45E-06	2	1.23E-05	5.78E-06
Fe	3	1.07E-01	4.84E-02	2	8.07E-02	9.15E-03	4	1.11E+00	9.51E-01
Fluoride	4	1.69E+00	6.42E-01	5	2.15E+00	6.89E-01	4	1.06E+00	3.86E-02
Gd	2	2.17E-05	1.43E-05	4	4.86E-06	3.84E-07	2	2.88E-05	1.97E-05
Hg	4	3.02E-03	1.01E-03	3	3.44E-03	8.60E-04	0	NA	NA
K	5	4.82E+00	1.19E+00	5	2.05E+01	1.78E+00	5	5.28E+00	4.05E-01
La	3	7.49E-05	3.97E-05	4	3.59E-05	3.91E-06	2	1.32E-04	8.91E-05
Li	3	2.70E-02	2.08E-02	4	6.91E-02	6.44E-03	1	7.75E-02	NA
Mg	5	8.66E+01	2.50E+01	5	1.37E+02	1.27E+01	6	1.13E+02	1.02E+01
Mn	4	1.68E-01	1.62E-01	5	7.25E-02	5.00E-02	4	2.61E-02	1.41E-02
Mo	5	1.95E-03	4.16E-04	5	3.72E-03	4.65E-04	6	2.29E-03	3.58E-04
Na	5	4.45E+02	1.42E+02	5	9.93E+02	9.93E+01	6	3.56E+02	2.02E+01
Nd	3	7.11E-05	4.44E-05	4	2.07E-05	3.71E-06	2	1.34E-04	9.66E-05
Ni	4	4.91E-03	1.39E-03	5	1.44E-02	4.35E-03	2	6.26E-03	1.66E-03
Nitrate	0	NA	NA	1	2.26E+00	NA	4	5.59E+00	8.11E-01
Pb	0	NA	NA	0	NA	NA	1	2.65E-03	NA
Phosphate	0	NA	NA	0	NA	NA	0	NA	NA
Pr	3	1.76E-05	9.70E-06	4	6.98E-06	5.57E-07	2	3.39E-05	2.28E-05
Sb	3	5.51E-04	1.39E-04	5	4.80E-04	5.80E-05	0	NA	NA
Se	3	3.67E-04	4.64E-05	3	8.68E-04	3.59E-05	5	4.82E-04	4.91E-05
Sm	4	2.77E-05	1.14E-05	4	3.24E-05	3.62E-06	2	3.91E-05	2.22E-05
Sn	0	NA	NA	0	NA	NA	0	NA	NA
Sr	5	3.91E+00	1.24E+00	5	7.39E+00	5.99E-01	6	4.59E+00	2.77E-01
Sulfate	4	1.55E+03	3.92E+02	5	2.27E+03	8.11E+01	6	1.29E+03	1.73E+02

Table continued on next page

**Table 17. Summary Statistics for Elemental Constituents in Surface Water Samples Collected in 1999 from Three Regional Reservoirs (Continued)**

Analyte	Concentration (mg L <sup>-1</sup> )								
	Brantley Lake			Red Bluff Reservoir			Lake Carlsbad		
	<sup>a</sup> N	<sup>b</sup> Mean	<sup>c</sup> SE	N	Mean	SE	N	Mean	SE
Th	3	1.30E-05	4.88E-06	3	1.56E-05	2.14E-06	2	3.77E-05	2.25E-05
Ti	4	3.31E-01	1.64E-01	4	6.58E-01	4.85E-02	2	4.57E-01	8.80E-02
Tl	0	NA	NA	0	NA	NA	0	NA	NA
U	4	2.42E-03	6.94E-04	4	6.50E-03	7.20E-04	2	6.48E-03	2.70E-03
V	5	3.82E-03	7.15E-04	5	3.96E-03	5.33E-04	6	6.09E-03	5.14E-04
Zn	1	1.70E-02	NA	1	1.79E-02	NA	2	1.59E-02	1.45E-03

<sup>a</sup>N = number of samples above MDL and included in calculations

<sup>b</sup>Mean = arithmetic mean

<sup>c</sup>SE = standard error of mean

<sup>d</sup>NA = not applicable; analyte detected in <2 samples

**Table 18. Summary Statistics for Elemental Constituents in Sediment Samples Collected in 1999 from Three Regional Reservoirs**

Analyte	Concentration (mg kg <sup>-1</sup> )								
	Brantley Lake			Red Bluff Reservoir			Lake Carlsbad		
	<sup>a</sup> N	<sup>b</sup> Mean	<sup>c</sup> SE	N	Mean	SE	N	Mean	SE
Ag	8	7.86E-02	3.94E-03	8	8.88E-02	4.35E-03	8	9.88E-02	5.71E-03
Al	12	2.23E+04	1.09E+03	12	1.37E+04	6.04E+02	12	9.06E+03	7.56E+02
As	12	4.55E+00	2.52E-01	12	4.76E+00	1.85E-01	12	3.31E+00	1.66E-01
Ba	12	2.04E+02	5.48E+00	12	2.62E+02	9.29E+00	12	1.40E+02	6.06E+00
Be	12	1.06E+00	7.84E-02	12	6.80E-01	4.17E-02	12	5.00E-01	4.99E-02
Ca	12	1.25E+05	9.09E+03	12	1.45E+05	9.54E+03	12	1.46E+05	1.07E+04
Cd	12	3.16E-01	2.99E-02	12	3.53E-01	3.86E-02	12	3.91E-01	4.98E-02
Ce	8	3.15E+01	1.98E+00	8	2.67E+01	1.80E+00	8	1.77E+01	1.44E+00
Chloride	12	5.39E+03	6.93E+02	12	1.10E+04	9.70E+02	12	3.04E+03	7.31E+02
Co	12	7.60E+00	2.97E-01	12	6.36E+00	2.78E-01	12	3.47E+00	2.49E-01
Cr	12	1.98E+01	1.07E+00	12	1.29E+01	4.89E-01	12	1.11E+01	7.24E-01
Cu	11	1.24E+01	1.46E+00	11	1.09E+01	1.61E+00	12	1.36E+01	2.07E+00
Dy	8	1.80E+00	1.63E-01	8	1.61E+00	1.40E-01	8	1.12E+00	1.01E-01
Er	8	8.48E-01	7.95E-02	8	7.91E-01	6.98E-02	8	5.56E-01	5.02E-02
Eu	8	6.88E-01	4.43E-02	8	5.79E-01	3.60E-02	8	3.86E-01	2.79E-02
Fe	12	1.81E+04	7.88E+02	12	1.34E+04	5.04E+02	12	9.20E+03	8.25E+02
Fluoride	0	<sup>d</sup> NA	NA	0	NA	NA	0	NA	NA
Gd	8	3.07E+00	2.33E-01	8	2.66E+00	1.94E-01	8	1.80E+00	1.47E-01
Hg	10	1.70E-02	9.92E-04	12	2.43E-02	1.25E-03	12	2.86E-02	2.30E-03
K	12	4.69E+03	2.11E+02	9	4.12E+03	2.21E+02	8	1.88E+03	1.63E+02

Table continued on next page

**Table 18. Summary Statistics for Elemental Constituents in Sediment Samples Collected in 1999 from Three Regional Reservoirs (Continued)**

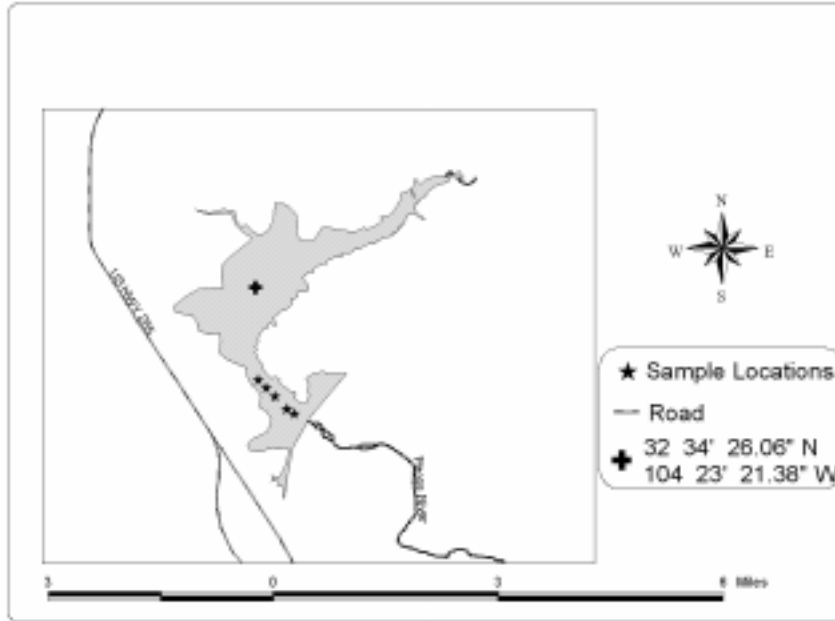
Analyte	Concentration (mg kg <sup>-1</sup> )								
	Brantley Lake			Red Bluff Reservoir			Lake Carlsbad		
	<sup>a</sup> N	<sup>b</sup> Mean	<sup>c</sup> SE	N	Mean	SE	N	Mean	SE
La	8	1.48E+01	8.03E-01	8	1.31E+01	6.34E-01	8	8.94E+00	6.33E-01
Li	8	2.50E+01	1.45E+00	8	1.52E+01	1.25E+00	8	1.16E+01	1.14E+00
Mg	12	1.56E+04	8.92E+02	12	1.05E+04	2.79E+02	12	1.30E+04	8.15E+02
Mn	12	4.65E+02	2.64E+01	12	4.29E+02	1.64E+01	12	2.92E+02	2.04E+01
Mo	12	1.06E+00	1.01E-01	12	1.85E+00	2.49E-01	8	3.81E-01	5.15E-02
Na	8	3.42E+03	3.52E+02	8	6.29E+03	3.78E+02	8	1.38E+03	1.37E+02
Nd	8	1.58E+01	9.79E-01	8	1.35E+01	6.57E-01	8	9.07E+00	6.68E-01
Ni	12	2.10E+01	1.18E+00	12	1.88E+01	1.49E+00	12	1.16E+01	1.15E+00
Nitrate	0	NA	NA	0	NA	NA	1	2.70E+00	NA
Pb	12	1.22E+01	8.79E-01	12	1.13E+01	1.02E+00	12	1.86E+01	3.12E+00
Phosphate	0	NA	NA	0	ND	ND	5	1.69E+01	4.67E+00
Pr	8	4.06E+00	2.21E-01	8	3.46E+00	1.72E-01	8	2.35E+00	1.72E-01
Sb	9	1.06E-01	1.78E-02	8	9.56E-02	6.82E-03	7	9.12E-02	1.69E-02
Se	12	1.68E+00	1.63E-01	12	2.46E+00	1.31E-01	12	1.77E+00	2.04E-01
Sm	8	3.21E+00	1.93E-01	8	2.71E+00	1.45E-01	8	1.84E+00	1.30E-01
Sn	0	NA	NA	0	NA	NA	0	NA	NA
Sr	12	4.83E+02	3.65E+01	12	6.03E+02	4.42E+01	12	5.34E+02	5.28E+01
Sulfate	12	5.98E+03	5.45E+02	12	8.35E+03	6.91E+02	12	5.75E+03	1.76E+03
Th	8	3.74E+00	4.35E-01	8	3.08E+00	3.31E-01	8	2.25E+00	2.97E-01
Ti	8	2.58E+02	2.34E+01	8	2.92E+02	7.14E+00	8	2.93E+02	1.97E+01
Tl	3	6.83E-01	5.49E-01	0	NA	NA	4	8.58E-01	3.71E-01
U	8	1.67E+00	1.49E-01	8	3.08E+00	3.32E-01	8	1.27E+00	1.11E-01
V	12	3.34E+01	1.47E+00	12	3.10E+01	1.96E+00	12	1.89E+01	1.03E+00
Zn	12	4.79E+01	3.59E+00	12	4.32E+01	4.17E+00	12	6.66E+01	8.01E+00

<sup>a</sup>N = number of samples above MDL and included in calculations

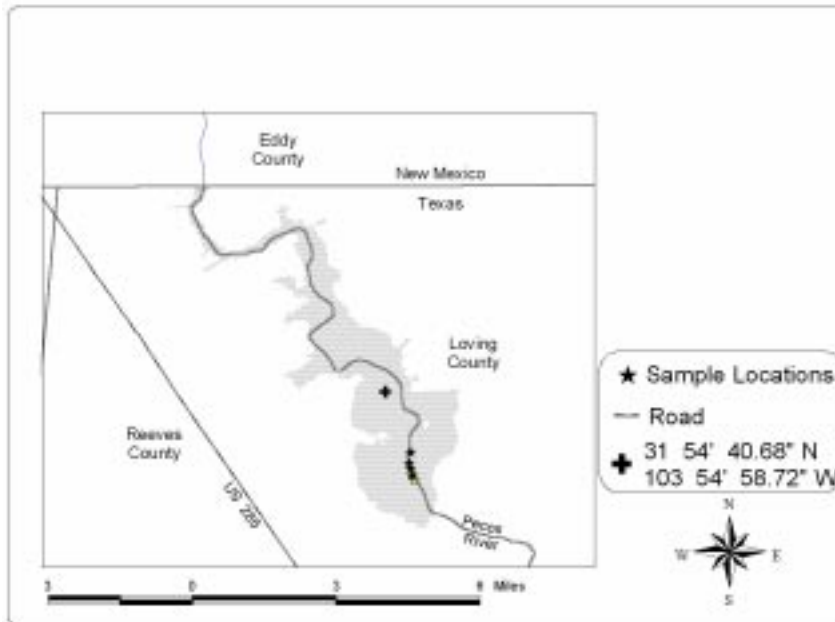
<sup>b</sup>Mean = arithmetic mean

<sup>c</sup>SE = standard error of mean

<sup>d</sup>NA = not applicable; analyte detected in <2 samples

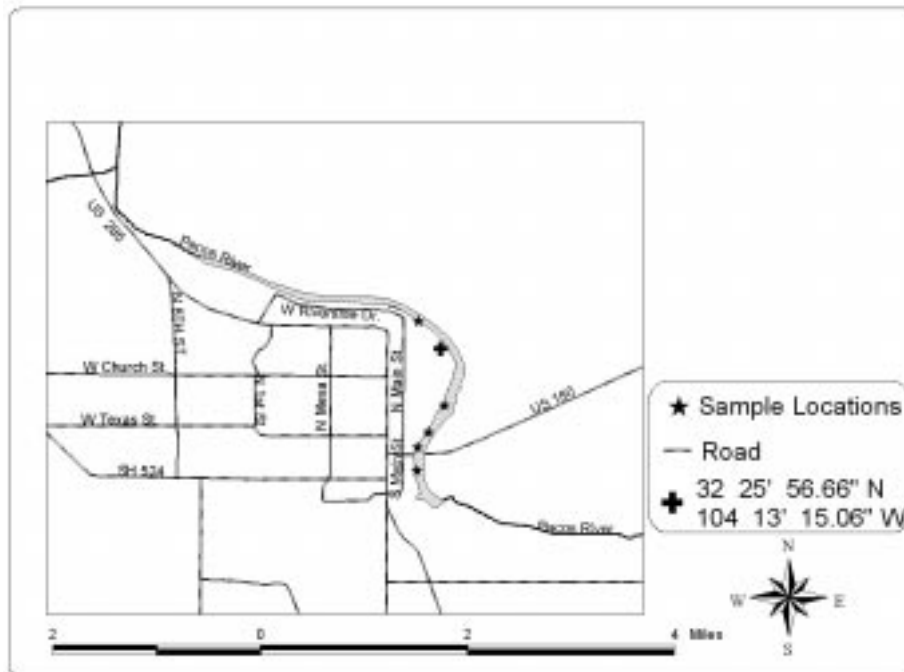


**Figure 23. Surface Water and Sediment Sampling Locations at Brantley Lake**

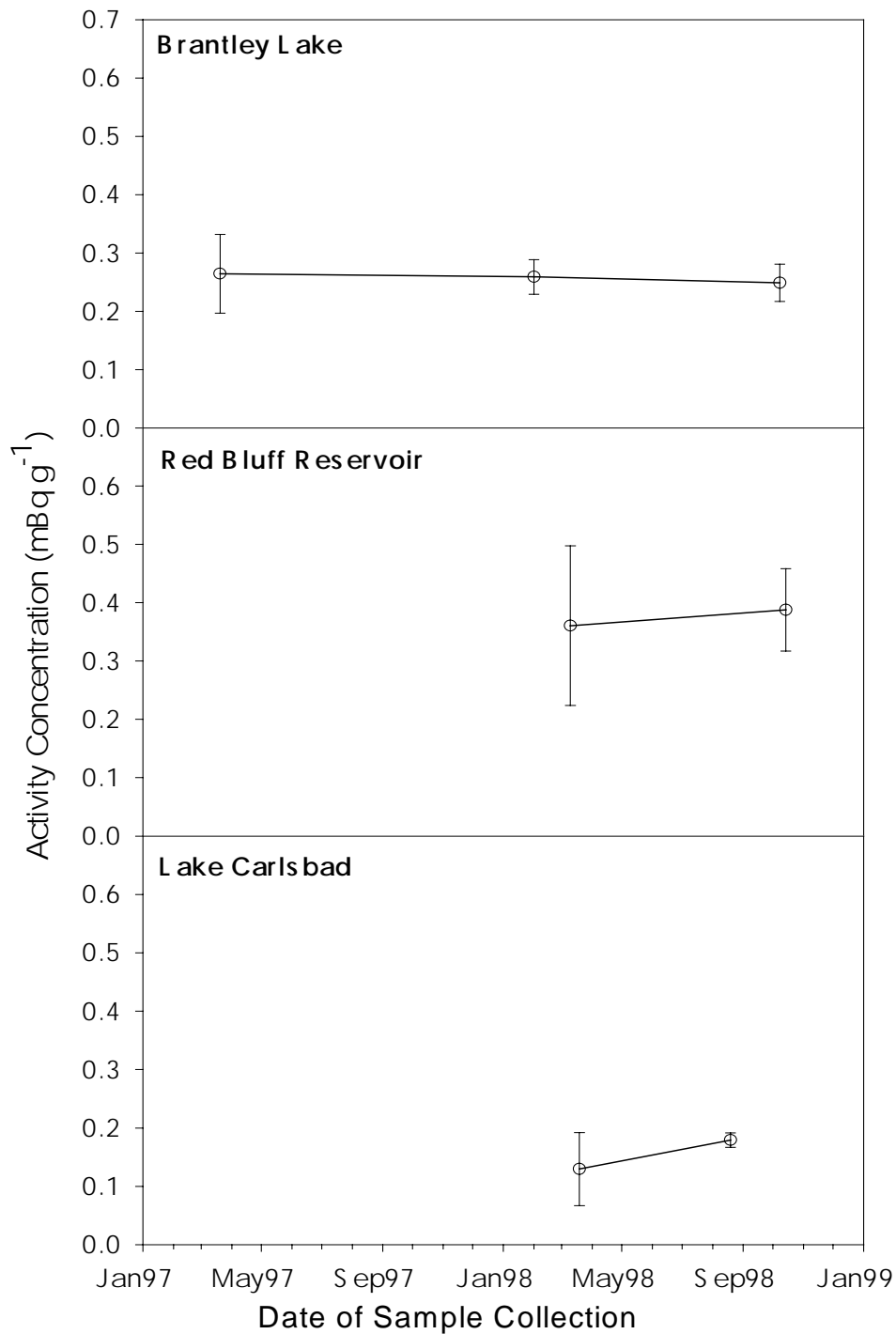


**Figure 24. Surface Water and Sediment Sampling Locations at Red Bluff Reservoir**



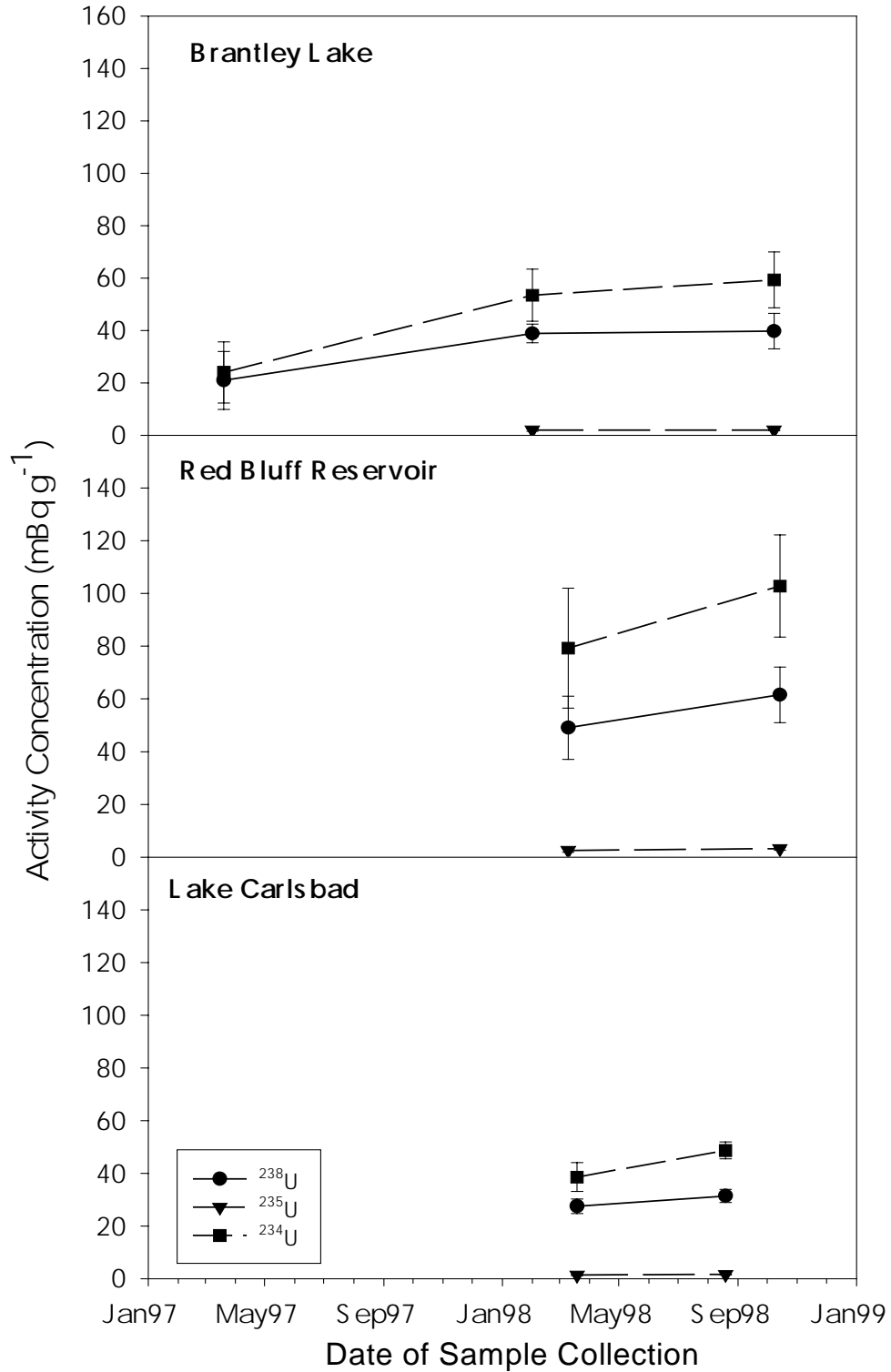


**Figure 25. Surface Water and Sediment Sampling Locations at Lake Carlsbad**



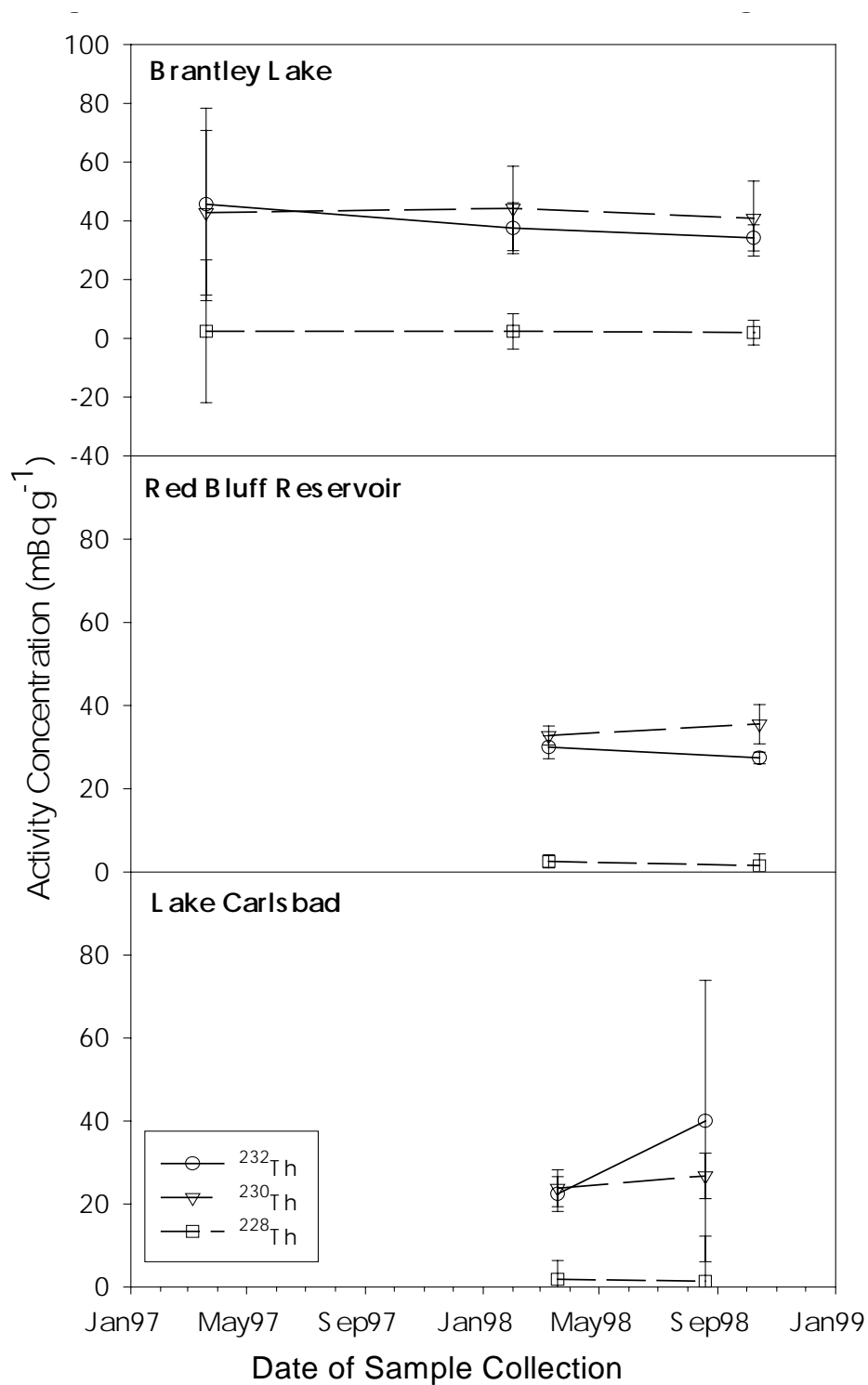
**Figure 26. Mean Baseline <sup>239,240</sup>Pu Activity Concentrations in Regional Reservoir Sediments Collected during 1997-1998**

Error bars represent 95% confidence interval of the mean.



**Figure 27. Mean Baseline U Isotope Activity Concentrations in Regional Reservoir Sediments Collected during 1997 - 1998**

Error bars represent 95% confidence interval of the mean.



**Figure 28. Mean Baseline Th Isotope Activity Concentrations in Regional Reservoir Sediments Collected during 1997 – 1998**

Error bars represent 95% confidence interval of the mean.

## Radiological and Non-radiological Constituents 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 July 1999, water samples were collected for CEMRC environmental monitoring studies from six sources in the region of the WIPP (Table 19).

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 source for the sampling site designated as Private Well #2 is a well seven miles southwest of the WIPP; this water is drawn from the Culebra aquifer.

The 1999 water samples were collected after WIPP started receiving radioactive waste. As of the date of this report, WIPP had not received any mixed waste. Therefore, this summary begins the CEMRC monitoring phase for radionuclides in drinking water but continues the baseline phase for non-radiological constituents. The baseline evaluation began in 1997, and results were reported in the CEMRC 1997 and 1998 reports. As in all of the other WIPP EM studies, the priorities of this work were to (1) continue establishing baseline concentrations for non-radiological constituents of environmental concern as a result of operations at the WIPP facility, (2) begin routine monitoring for radionuclides of concern, (3) continue providing data useful for exposure modeling and (4) refine analytical techniques to improve data quality.

In 1997, drinking water samples were collected for radioanalytical and related studies from the sources described above (the 1997 sample from the Double Eagle well field was

collected at a private tap). In 1998, a 200-L sample for radiological analyses and a 9-L sample for non-radiological analyses were collected from each of the six sites sampled in 1999.

Actinide analyses on ~100-L drinking water samples were conducted by CEMRC in 1997 using chemical separation and alpha spectroscopy methods. The results indicated that  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  were not present at levels greater than the MDCs of  $5.3 \mu\text{Bq L}^{-1}$  and  $8.8 \mu\text{Bq L}^{-1}$ , respectively. Even though Pu and Am would present no significant health hazard at levels less than these MDCs, it was desirable to quantify the levels in order to provide better identification of any changes after the WIPP began receiving waste. For these ultra-low level studies, the 200-L samples collected in 1998 and four additional 200-L samples collected in 1999 were sent to Los Alamos National Laboratory for thermal ionization mass spectrometry (TIMS) analyses for  $^{239}\text{Pu}$ . Three liters of each 1998 drinking water sample were also saved in Marinelli beakers for additional radionuclide analyses by CEMRC.

The NMSU SWAT laboratory analyzed drinking water samples collected in 1997 and 1998 for non-radiological constituents. CEMRC also analyzed 1998 samples and the results indicated a good agreement between the two laboratories. Drinking water from the Carlsbad region has a high mineral content, but the overwhelming majority of inorganic analytes were at concentrations well below Safe Drinking Water Act (SDWA) reference levels. Some metals of concern, due to their expected occurrence in WIPP mixed waste, (notably Be, Hg and Ag) were not detected by SWAT in the 1997 nor the 1998 samples. These elements, however, were detected in a few of the 1998 samples analyzed by CEMRC using a newly acquired ICP-MS. Five other metals, (Sr, Bi, Co, Mn, Mo and V) were also quantified by CEMRC in 1998 samples, but they were not detected by SWAT. SWAT detected organic analytes in a few of the 1998

samples, (dibromochloromethane and bromoform) but at levels below SDWA limits.

In summary, the CEMRC analyses of drinking water samples collected in 1998 were consistent with the results of the previous year's analyses based on SWAT data. Few organic contaminants were detected and those inorganic substances that were quantified were, with a few exceptions, below SDWA reference levels. With the exception of Se analyses and variations possibly associated with sample preservation, the CEMRC data appear to be comparable with results from the SWAT laboratory.

## Methods

All 1999 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 metals 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 1999 samples were collected at the same six locations as the 1998 samples.

CEMRC performed the non-radiological analyses using IC, ICP-MS and AAS. Instrumentation, general methods and quality assurance (QA) work are presented in Appendix K. CEMRC did not test the 1999 drinking water samples for organic constituents because of low and consistent results in prior studies.

Radiological analyses were carried out at CEMRC by first counting the samples in Marinelli beakers using a coaxial, HPGe detector system to determine gamma-emitting radionuclide concentrations. The water was then transferred to the radiochemistry laboratory where actinides were separated and purified using multiple precipitation, co-precipitation and ion-exchange and/or extraction chromatography. Once the actinides were separated elementally, they were co-precipitated with La and deposited onto filters, which were then counted on an alpha spectroscopy system. Radioanalytical QA/QC data are presented in Appendix L.

TIMS analyses were applied to 200-L samples of water from each source that were collected separately from samples analyzed at

CEMRC. TIMS analyses also used multiple precipitation, co-precipitation and ion-exchange and/or extraction chromatography techniques, but the purified Pu was co-plated with Pt onto a Re thermal ionization mass spectrometry filament. A thin layer of Pt was deposited over the Pu-Re layer to form a surface ionization diffusion controlled source for mass spectrometric analyses.

## Results and Discussion

### Radiological

No radionuclides were measured above MDC in 1998 or 1999 samples as measured by gamma spectroscopy targeting 11 naturally occurring and 12 anthropogenic gamma-emitters. This was expected due to the small sample size used for this screening effort. Naturally occurring actinides were detected in all of the samples from each location in each of the two years. The greatest concentrations of  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{234}\text{U}$  and  $^{228}\text{Th}$  were detected in Private Well #2. The sum of measured actinide concentrations for the 1999 sample from this well was  $743 \text{ mBq L}^{-1}$ . It was followed by Otis ( $207 \text{ mBq L}^{-1}$ ), Hobbs ( $128 \text{ mBq L}^{-1}$ ), Loving ( $113 \text{ mBq L}^{-1}$ ), Double Eagle ( $87 \text{ mBq L}^{-1}$ ) and Carlsbad ( $42 \text{ mBq L}^{-1}$ ).

The uranium isotope concentrations in 1999 water samples were quite similar ( $\pm 16\%$ ) to the 1998 samples.  $^{234}\text{U}$  concentrations were 2.6-3.2 times greater than  $^{238}\text{U}$  concentrations as was expected. Progeny radionuclides, ( $^{234}\text{Th}$ ,  $^{234\text{m}}\text{Pa}$ ,  $^{234}\text{U}$  and  $^{230}\text{Th}$ ) are believed to recoil into the water from the alpha decay of the parent,  $^{238}\text{U}$ . There they may be suspended or dissolved and transported with the water within the aquifer.  $^{228}\text{Th}$  (a progeny of  $^{232}\text{Th}$ , which was not detected) varied considerably between the two years, especially at Otis (350% higher in the 1999 sample). Th is somewhat less soluble in water than U and thus may be less susceptible to transport. This helps to explain why U concentrations were always greater than Th concentrations, most of which were not detected even though the parent isotopes ( $^{238}\text{U}$  and  $^{232}\text{Th}$ ) are expected to be in equal radiological abundance (Taylor, S. R. and S. M. McLennan, 1995, Rev. of Geophys 33,241).

Pu was not detected in 1999 or 1998 drinking water samples processed by actinide separation chemistry and alpha spectroscopy. A few individual results initially indicated that  $^{239}\text{Pu}$  might be present, but these results were determined to be due to laboratory artifacts, and no Pu was observed in a second set of samples. Again, TIMS analyses on drinking water samples did not indicate  $^{239}\text{Pu}$  was present above the maximum MDC of  $\sim 3 \mu\text{Bq L}^{-1}$  (Table 19) which is approximately ten times lower than minimum MDCs of CEMRC analyses using 3-L samples. One may summarize, therefore, that Pu has still not been detected in drinking water from the local area. Finally, the radioanalytical results for U agreed quite favorably with the results obtained by ICP-MS collected in both 1998 and 1999. In contrast, the Th results were quite variable, possibly as a result of differences in solubility of U and Th.

### **Non-Radiological Results**

To evaluate trends in concentrations over time, the concentration data for drinking water samples collected in 1998 and 1999 were compared. The concentrations of almost all analytes in the sample pairs were in close agreement (Table 20). Of the 264 pairs of values compared, there were only two cases in which the differences between years were a factor of 5 or greater.

There were no particularly noteworthy differences in the Loving or Otis samples for 1997 as compared with 1998 and 1999 samples, except for a higher Sb level in the sample from Loving. As in 1998, several inorganic, non-radiological substances exceeded reference levels (secondary maximum contaminant levels) in the 1999 samples from the Otis and Private Well #2 sources. Specifically, these were chloride (by autoanalyzer), and sulfate. The fact that the 1997, 1998 and 1999 data showed essentially identical patterns for these two analytes indicates that the high concentrations were not spurious results but rather a true indication of elevated levels.

With respect to heavy metals, the Cr level in the 1999 Carlsbad sample was 793% greater than in the 1998 sample with an observed concentration of  $2.7 \mu\text{g L}^{-1}$ . For the Hobbs samples, the Pb concentration was a factor of 44

lower in the August 1998 sample compared with the July 1997 sample ( $0.2$  vs.  $8.7 \mu\text{g L}^{-1}$ ), and even lower in the 1999 sample ( $0.09 \mu\text{g L}^{-1}$ ). In contrast, the Tl concentration was more than four-fold higher in the 1998 Hobbs sample compared to the 1997 sample ( $0.26$  vs.  $0.05 \mu\text{g L}^{-1}$ ), but was non-detectable in the 1999 sample. The greatest Pb levels were observed in samples from Loving ( $1.7 \mu\text{Bq L}^{-1}$ ), Carlsbad ( $1.4 \mu\text{g L}^{-1}$ ) and Private Well #2 ( $1.4 \mu\text{g L}^{-1}$ ). The Cu and Pb concentrations were well below SDWA reference levels in all samples. As was the case in 1997, the highest concentrations of Ni and Tl were found in drinking water from Private Well #2, and as before, those concentrations were well below reference levels. In fact, the concentrations of Ni, Mg, Co, As and Al were lower at all sites in 1999, and Se levels were below the MDC of  $0.12 \mu\text{g L}^{-1}$ .

It is important to emphasize that these results are not appropriate for use in assessing of regulatory compliance. However, it is noteworthy that the CEMRC results for Carlsbad drinking water collected in 1999 agreed very well with the measurements published by the City of Carlsbad Water Department (*1998 Annual Consumer Report on the Quality of Tap Water*). All of the CEMRC values were within the range of concentrations reported by the City of Carlsbad except for nitrate. CEMRC reported levels of  $4.9$ - $5.9 \text{ mg L}^{-1}$  of nitrate in 1998 and 1999 samples, while Carlsbad reported  $0.81 \text{ mg L}^{-1}$  for a single sample collected in November of 1998. City of Carlsbad samples were collected at wellheads, while CEMRC samples were collected at a downstream point where water from individual wells has been combined, which may have contributed to the differences in nitrate measurements. However, the  $\sim 5 \text{ mg L}^{-1}$  is still less than half of the EPA maximum contaminant level of 10 ppm. Tables presenting drinking water data summarized herein are available on the CEMRC web site at <http://www.cemrc.org>.

**Table 19.  $^{239}\text{Pu}$  Activity Concentrations in Drinking Water Samples Collected near WIPP as Determined by Thermal Ionization Mass Spectrometry**

Drinking Water Sample			$^{239}\text{Pu}$ Activity Concentration (nBq L <sup>-1</sup> )	MDC (nBq L <sup>-1</sup> )
Location	Date Collected	Weight (kg)		
Otis	28 October 1998	218	< <sup>a</sup> MDC	1,300
Carlsbad	21 October 1998	216	<MDC	3,200
Double Eagle	23 February 1999	219	<MDC	172
Loving	22 February 1999	220	<MDC	172
Private Well #2	5 April 1999	220	<MDC	172
Hobbs	23 August 1999	220	<MDC	172

<sup>a</sup>MDC = minimum detectable concentration**Table 20. Difference between Concentrations of Selected Analytes in Drinking Water Samples Collected during 1998 and 1999**

Analyte	<sup>a</sup> Percent Difference by Location					
	Carlsbad	Double Eagle	Hobbs	Private Well #2	Otis	Loving
Al	-86	-32	-55	-50	<sup>b</sup> ND	-30
As	-2	-16	-14	-10	-18	-27
Ba	9	29	12	2	-10	9
Ca	-24	-12	-13	5	-3	-15
Co	ND	-30	-27	-16	ND	-72
Cr	793	-16	8	8	261	50
Cu	ND	108	9	77	ND	93
Fe	ND	-53	-35	19	ND	ND
K	-25	20	14	0	24	24
Mg	-12	-1	-2	-14	-6	-13
Mo	73	-36	-3	-12	ND	-5
Na	-58	-15	-19	-22	17	-20
Ni	ND	-31	-36	-23	ND	-15
Pb	ND	101	-45	44	ND	19
Sb	-93	-97	ND	ND	ND	704
Sr	-21	6	13	23	2	26
Tl	ND	ND	ND	-40	ND	ND
<sup>228</sup> Th	-62	-33	44	-34	349	-18
<sup>238</sup> U	-16	14	3	-1	14	9
<sup>235</sup> U	-7	2	14	-7	4	-9
<sup>234</sup> U	-12	12	-3	0	16	4
V	17	-33	-35	-35	3	-24
Zn	230	86	65	-45	ND	64

Table continued on next page



**Table 20. Difference between Concentrations of Selected Analytes in Drinking Water Samples Collected during 1998 and 1999 (Continued)**

Analyte	<sup>a</sup> Percent Difference by Location					
	Carlsbad	Double Eagle	Hobbs	Private Well #2	Otis	Loving
Fluoride	-38	-13	-12	-20	-41	-5
Sulfate	-2	-10	-21	8	15	3
Nitrate	-18	-6	-9	3	15	-9
Chloride	-58	-5	-24	-5	42	-21

<sup>a</sup>Percent Difference = (1999 concentration – 1998 concentration) / 1998 concentration \* 100%

<sup>b</sup>ND = analyte not detected in one or both samples

## Occurrence of Radionuclides in Residents of the Carlsbad, New Mexico Area

### 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 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 1999) 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, instrument and detection limits is provided in the CEMRC 1998 Report. In addition, the status of project and results are posted monthly on the CEMRC website (<http://www.cemrc.org>) and semi-annually through a newsletter distributed throughout the local community.

### Results

As of 1 October 1999, 412 individuals have 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 of the original cohort began in July 1999, and 23 recounts had been performed through 1 October 1999.

Demographic characteristics (Table 21) of the current LDBC cohort are generally consistent with those reported in the CEMRC

1998 Report and the 1990 census for citizens living in Carlsbad. The largest deviations reported in the 1998 report between the LDBC cohort and 1990 census were the over-sampling of males and under-sampling of Hispanics. Since that time, improvements have been made with respect to the percent of males sampled, but Hispanics are still over 50% under-sampled relative to the 1990 census. In the future, demographic characteristics of the LDBC project will be compared to results of the 2000 census, which will be 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.

As discussed in detail in the CEMRC 1998 Report, 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 pairwise 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  $^{232}\text{Th}$  via  $^{212}\text{Pb}$ ,  $^{235}\text{U}$  /  $^{226}\text{Ra}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ ,  $^{54}\text{Mn}$ ,  $^{232}\text{Th}$  via  $^{228}\text{Ac}$  and  $^{65}\text{Zn}$  (Table 22). As discussed in detail in the 1998 report, five of these ( $^{232}\text{Th}$  via  $^{212}\text{Pb}$ ,  $^{60}\text{Co}$ ,  $^{40}\text{K}$ ,  $^{54}\text{Mn}$  ( $^{228}\text{Ac}$  interference) and  $^{232}\text{Th}$  via  $^{228}\text{Ac}$ ) are part of the shield-room background and positive detection is expected at low frequency.  $^{40}\text{K}$  is a naturally occurring isotope of an essential biological element, so detection in all individuals is expected.  $^{137}\text{Cs}$  and  $^{235}\text{U}$  /  $^{226}\text{Ra}$  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). It is interesting that no result above  $L_C$  has been observed for  $^{65}\text{Zn}$ . This finding may be a statistical anom-

aly, but may suggest an abnormality in the analysis algorithm at that photon energy.

For the operational monitoring counts (Table 22,  $n = 62$ ), the percentage of results greater than  $L_C$  were consistent with baseline at a 95% confidence level (margin of error). The margin of error could not be calculated for many of the percentages because the sample size of the operational monitoring counts was too small at the time of this summary. When sufficient operational monitoring counts are performed, the margin of error will be calculated. In addition, 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 a change 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.

$^{40}\text{K}$  results were positive for all participants ( $n = 412$ ), ranging from 2120 to 6504 Bq per person with an overall mean ( $\pm$  SE) of 3959 ( $\pm$  43) Bq. 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  $^{40}\text{K}$ . The mean  $^{40}\text{K}$  value for males ( $\pm$ SE), was 4531 ( $\pm$  43) Bq, which was significantly greater ( $P < 0.0001$ ) than that of females, which was 3218 ( $\pm$  38) Bq. This result was expected since, in general, males tend to have larger body sizes and greater muscle content than females.

Detectable  $^{137}\text{Cs}$  is present in  $27.0 \pm 4.4$  % (95% confidence level, baseline and operational monitoring counts) of citizens living in the Carlsbad area. These results are consistent with preliminary findings reported in the CEMRC 1997 and 1998 reports. Detectable  $^{137}\text{Cs}$  body burdens ranged from 5.6 to 62.9 Bq with an overall mean ( $\pm$  SE) of 10.8 ( $\pm$  0.6) Bq. The mean  $^{137}\text{Cs}$  body burden for males ( $\pm$ SE), was 11.6 ( $\pm$  0.9) Bq, which was significantly greater ( $p = 0.01$ ) than that of females, which were 9.0 ( $\pm$  0.5 Bq).

Individual  $^{137}\text{Cs}$  results were compared to two parameters of demographic data to deter-

mine whether the presence of  $^{137}\text{Cs}$  was dependent on a particular demographic or lifestyle characteristic, using a Chi-square test of independence (Table 23). The presence of  $^{137}\text{Cs}$  was independent of ethnicity, age, radiation work history, consumption of wild game, nuclear medical treatments and European travel. Occurrence of detectable  $^{137}\text{Cs}$  was significantly associated ( $p = 0.003$ ) with gender, where males had higher prevalence of  $^{137}\text{Cs}$  relative to females. Presence of  $^{137}\text{Cs}$  was also associated with smoking habit (where smokers had a higher prevalence relative to non-smokers), but at a lower significant level ( $p=0.032$ ). The results reported herein are consistent with those reported in the CEMRC 1998 Report, except that gender association was not significant ( $p = 0.06$ ) in the earlier subset of the baseline data. The frequency of smoking among males and females was statistically indistinguishable (17.8 % for males, 16.6 % for females,  $p = 0.807$ ), eliminating a correlation between the two variables as a confounding factor. 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  $^{40}\text{K}$  results. The association of  $^{137}\text{Cs}$  with smoking habit could be related to the presence of fallout  $^{137}\text{Cs}$  in tobacco, decreased pulmonary clearing capability in smokers, or other as yet unidentified factors, and further study is warranted.

As reported in the CEMRC 1998 Report, the percentage of results greater than  $L_C$  for  $^{235}\text{U}/^{226}\text{Ra}$  (11.1%) 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  $^{137}\text{Cs}$ , but the large sample size of the current cohort tends to support the observed pattern. Although  $^{235}\text{U}$  and  $^{226}\text{Ra}$  cannot be differentiated via gamma spectroscopy, it is likely the signal is the result of  $^{226}\text{Ra}$  because the natural abundance of  $^{226}\text{Ra}$  is much greater than that of  $^{235}\text{U}$ . However, further study, possibly involving *in vitro* bioassay for the two radionuclides (the nuclides can be distinguished via alpha spectrometry), is needed.

**Table 21. Demographic Characteristics of the "Lie Down and Be Counted" (LDBC) Population Sample through 1 October 1999**

Characteristic		LDBC, 1999 ( <sup>a</sup> margin of error)	LDBC, 1998	<sup>b</sup> Census, 1990
Gender	Male	56.6% (52.2 to 61.9%)	60.7%	48.0%
	Female	43.43% (38.6 to 48.3%)	39.3%	52.0%
Ethnicity	Hispanic	12.9% (9.5 to 16.3%)	14.0%	33.4%
	Non-Hispanic	84.2% (80.6 to 87.9%)	82.7%	63.0%
	Other	2.9% (1.2 to 4.6%)	3.3%	3.6%
Age 60 or older		24.8% (20.3 to 29.1%)	25.7%	33.7%
Currently or previously classified as a radiation worker		4.9% (2.7 to 7.0%)	4.0%	<sup>c</sup> NA
Consumption of wild game within last 3 months		15.3% (11.7 to 18.9%)	16.9%	NA
Medical treatment, other than x-rays, using radionuclides		9.0% (6.1 to 11.9%)	8.5%	NA
European travel within the last 2 years		3.9% (1.9 to 5.8%)	4.4%	NA
Current smoker		14.6% (10.9 to 18.2%)	14.0%	NA

<sup>a</sup>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.

<sup>b</sup>United States Department of Commerce, Economics and Statistics Administration, Bureau of the Census. *1990 Census of Population*. Washington, DC: US Government Printing Office

<sup>c</sup>NA = not available

**Table 22. Lie Down and Be Counted Results through 1 October 1999**

Radionuclide	In Vivo Count Type	Baseline Counts (prior to 27 March 1999) <sup>a</sup> N = 367	Operational Monitoring Counts (27 March 1999 – 1 October 1999) N = 62	
		% of Results ≥ <sup>b</sup> L <sub>C</sub>	% of Results ≥ <sup>b</sup> L <sub>C</sub>	<sup>c</sup> Margin of Error (%)
<sup>241</sup> Am	Lung	5	8	0 to 18
<sup>144</sup> Ce	Lung	5	2	<sup>d</sup> NC
<sup>252</sup> Cf	Lung	4	10	0 to 18
<sup>244</sup> Cm	Lung	6	6	0 to 18
<sup>155</sup> Eu	Lung	7	5	NC
<sup>237</sup> Np	Lung	4	3	NC
<sup>210</sup> Pb	Lung	4	8	0 to 18
Plutonium Isotope	Lung	6	3	NC
<sup>e232</sup> Th via <sup>212</sup> Pb	Lung	34	44	33 to 64
<sup>232</sup> Th	Lung	5	6	NC
<sup>232</sup> Th via <sup>228</sup> Th	Lung	4	6	0 to 18
<sup>233</sup> U	Lung	6	15	4 to 31
<sup>235</sup> U / <sup>226</sup> Ra	Lung	11	11	4 to 31
Natural Uranium via <sup>234</sup> Th	Lung	5	8	0 to 18
<sup>133</sup> Ba	Whole Body	4	8	0 to 18
<sup>140</sup> Ba	Whole Body	5	2	NC
<sup>141</sup> Ce	Whole Body	4	3	NC
<sup>58</sup> Co	Whole Body	4	3	NC
<sup>e60</sup> Co	Whole Body	55	44	29 to 60
<sup>51</sup> Cr	Whole Body	6	5	0 to 18
<sup>134</sup> Cs	Whole Body	2	5	0 to 18
<sup>137</sup> Cs	Whole Body	28	15	4 to 31
<sup>152</sup> Eu	Whole Body	7	2	NC
<sup>154</sup> Eu	Whole Body	4	3	NC
<sup>155</sup> Eu	Whole Body	4	2	<sup>d</sup> NC
<sup>59</sup> Fe	Whole Body	4	3	<sup>d</sup> NC
<sup>131</sup> I	Whole Body	5	2	<sup>d</sup> NC
<sup>133</sup> I	Whole Body	3	5	<sup>d</sup> NC
<sup>192</sup> Ir	Whole Body	4	3	<sup>d</sup> NC
<sup>40</sup> K	Whole Body	100	100	<sup>d</sup> NC
<sup>54</sup> Mn	Whole Body	<sup>c</sup> 12	<sup>c</sup> 13	0 to 18
<sup>103</sup> Ru	Whole Body	2	2	<sup>d</sup> NC
<sup>106</sup> Ru	Whole Body	4	2	<sup>d</sup> NC
<sup>125</sup> Sb	Whole Body	5	3	<sup>d</sup> NC

Table continued on next page

**Table 22. Lie Down and Be Counted Results through 1 October 1999  
(Continued)**

Radionuclide	<i>In Vivo</i> Count Type	Baseline Counts (prior to 27 March 1999) <sup>a</sup> N = 367	Operational Monitoring Counts (27 March 1999 – 1 October 1999) N = 62	
		% of Results ≥ <sup>b</sup> L <sub>C</sub>	% of Results ≥ <sup>b</sup> L <sub>C</sub>	<sup>c</sup> Margin of Error (%)
<sup>232</sup> Th via <sup>228</sup> Ac	Whole Body	<sup>c</sup> 35	<sup>c</sup> 24	13 to 40
<sup>88</sup> Y	Whole Body	8	3	<sup>d</sup> NC
<sup>65</sup> Zn	Whole Body	0	0	<sup>d</sup> NC
<sup>95</sup> Zr	Whole Body	7	5	<sup>d</sup> NC

<sup>a</sup>N = number of individuals

<sup>b</sup>To determine whether or not activity has been detected in a particular person, the parameter L<sub>C</sub> is used; the L<sub>C</sub> represents the 95<sup>th</sup> 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<sub>C</sub>

<sup>c</sup>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

<sup>d</sup>Not Calculated; the margin of error cannot be calculated for the observed percentage because the sample size for operational monitoring counts is as yet too small; when sufficient counts are performed, the margin of error will be calculated

<sup>e</sup>These radionuclides are present in the shield background, so they are expected to be detected periodically

**Table 23. Demographic Characteristics Associated with Occurrence of  $^{137}\text{Cs}$  in Local Residents**

Characteristic		Percentage of Population ( <sup>a</sup> N=300) where $^{137}\text{Cs}$ was Not Detected ( $H_0$ ), ( <sup>b</sup> margin of error)	Percentage of Population ( <sup>a</sup> N =111) where $^{137}\text{Cs}$ was Detected ( $H_A$ ), ( <sup>b</sup> margin of error)	<sup>c</sup> p-value ( $H_A \subset H_0$ )
Gender	Male	52.0 (46.3 to 57.7)	68.5 (59.5 to 77.5)	0.003
	Female	48.0 (42.3 to 53.7)	31.5 (22.5 to 40.5)	
Ethnicity	Hispanic	12.7 (8.7 to 16.7)	13.5 (6.3 to 20.7)	0.319
	Non-Hispanic	84.7 (80.3 to 89.0)	82.9 (75.7 to 90.1)	
	Other	2.6 (0.7 to 4.7)	3.6 (0.0 to 7.2)	
Age	≥ 60 years	25.0 (20.0 to 30.0)	24.3 (16.2 to 32.4)	0.888
	< 60 years	75.0 (70.0 to 80.0)	75.7 (67.6 to 83.8)	
Currently or previously classified as a radiation worker	Yes	5.7 (3.0 to 8.3)	2.7 (0.0 to 8.1)	0.215
	No	94.3 (91.7 to 97.0)	97.3 (91.7 to 100.0)	
Consumption of wild game within last 3 months	Yes	15.0 (10.7 to 19.3)	16.2 (9.0 to 23.4)	0.761
	No	85.0 (80.7 to 89.3)	83.8 (76.6 to 91.0)	
Medical treatment, other than x-rays, using radionuclides	Yes	7.7 (4.3 to 11.0)	12.6 (6.3 to 18.9)	0.120
	No	92.3 (89.0 to 95.7)	87.4 (81.1 to 93.7)	
European travel within the last 2 years	Yes	3.3 (1.0 to 5.7)	5.4 (0.9 to 9.9)	0.335
	No	96.7 (94.3 to 99.0)	94.6 (89.2 to 100.0)	
Current smoker	Yes	12.3 (8.3 to 16.0)	20.7 (13.5 to 28.8)	0.033
	No	87.7 (83.7 to 91.3)	79.3 (72.1 to 87.4)	

<sup>a</sup>N = number of individuals

<sup>b</sup>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

<sup>c</sup>The probability of observing a percentage greater than  $H_A$  assuming that the percentage ( $H_0$ ) is the true value





## **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, New Mexico, 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.

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 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-55<sup>th</sup> District). On 23 March 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. However, CEMRC continues to conduct various collaborative activities with WERC.

**Appendix B. Subcontractors for Scientific Work during 1999**

<b>Subcontractor</b>	<b>Scope of Work</b>
Battelle Memorial Institute, Pacific Northwest Division	Fabrication of lung sets for <i>in vivo</i> bioassay
Duke Engineering and Services	Radioanalyses of sediment samples
Electronic Counter Corporation	Instrument design & engineering
James Neton, Ph.D.	Assessment of <i>in vivo</i> radiobioassay program
Los Alamos National Laboratory	Analyses of water samples by thermal ionization mass spectrometry
National Institute of Standards & Technology	Intercomparison services for radioanalyses
NMSU Soil, Water, Air Testing Laboratory	Analyses of soil samples
Oak Ridge National Laboratory, Intercomparison Studies Program	Intercomparison services for <i>in vivo</i> radiobioassay
University of Rhode Island/Urszula Tomza	Neutron activation analysis, gamma-ray spectroscopy

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

Member/Term of Service	Affiliation
Stanley I. Auerbach, Ph.D. (PRB) / 1998-present	Director Emeritus, Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee
John M. Briggs, Ph.D. (SAB) / 2000-2001	Associate Professor, Department of Plant Biology, Arizona State University, Tempe, Arizona
O. Doyle Markham, Ph.D. (PRB) / 1998-present	Director, Environmental Science & Research Foundation, Inc., Idaho Falls, Idaho
Michael H. Smith, Ph.D. (PRB) / 1998-present	Director, Savannah River Ecology Laboratory, University of Georgia, Aiken, South Carolina
Thomas A. Cahill, Ph.D. (SAB) / 1998-1999	Professor (Emeritus), Atmospheric Sciences/Physics, University of California, Davis, Davis, California
Milan S. Gadd, Ph.D. (SAB) / 1998-1999	Senior Health Physicist, Rocky Flats Environmental Technology Site, Golden, Colorado
Shawki A. Ibrahim, Ph.D. (SAB) / 2000-2001	Professor, Department of Radiological Health Sciences, Colorado State University, Ft. Collins, Colorado
Kenneth G.W. Inn, Ph.D. (SAB) / 1998-1999	Group Leader, Office of Radiation Measurements, Ionizing Radiation Division, National Institute of Standards and Technology, Washington, D.C.
William K. Michener, Ph.D. (SAB) / 1998-1999	Associate Scientist, J.W. Jones Ecological Research Center, Newton, Georgia
F. Ward Whicker, Ph.D. (SAB) / 1998-1999	Professor and Department Head, Department of Radiological Health Sciences, Colorado State University, Ft. Collins, Colorado

### Appendix D. Professional Presentations and Publications during 1999

Author	Title	Publisher/Conference
Arimoto, R.	Sources and composition of aerosol particles	Handbook of Atmospheric Chemistry, submitted
Arimoto, R.	Eolian dust and climate: relationships to sources, transport, and deposition	Earth Science Reviews, submitted
Arimoto, R., J.A. Snow, W.C. Graustein, J.L. Moody, B.J. Ray, R.A. Duce, K.K. Turekian and H.B. Maring	Influences of atmospheric transport pathways on radionuclide activities in aerosol particles from over the North Atlantic	Journal of Geophysical Research 104:21301-21316
Arimoto, R.	Mineral aerosols, tropospheric chemistry, and climate forcing	Loessfest '99, Bonn, Germany
Arimoto, R., A.M. Nottingham, and C. Schloesslin	Aerosol sulfate and other aerosol constituents at the South Pole	American Geophysical Union, Fall Meeting, San Francisco, California
Chen, G., J. H. Crawford, D. D. Davis, F. L. Eisele, R. L. Mauldin, D. J. Tanner, M. D. Buhr, J. B. Nowak, R. Arimoto, S. Liu, P. Wang, X. Gong, B. J. DiNunno, B. L. Lefer, and R. E. Shetter	A comparison of model results with observations recorded during project ISCAT	American Geophysical Union, Fall Meeting, San Francisco, California
Conley, M.	Overview of WIPP Environmental Monitoring project	Meeting of Committee on the WIPP, National Academy of Science, National Academy of Engineering and Institute of Medicine Meeting, Albuquerque, New Mexico
Guelle, W., Y. Balkanski, M. Schulz, B. Marticorena, G. Bergametti, C. Moulin, R. Arimoto and K.D. Perry	Modeling the atmospheric distribution of mineral aerosol: comparison with ground measurements and satellite observations for yearly and synoptic time scales over the North Atlantic	Journal of Geophysical Research, in press
Huang, S.R., R. Arimoto, and K.A. Rahn	Sources and source variations for aerosol at Mace Head, Ireland	American Geophysical Union, Fall Meeting, San Francisco, California
Huang, S., K. A. Rahn, and R. Arimoto	Testing and optimizing two factor-analysis techniques on aerosol at Narragansett, Rhode Island	Atmospheric Environment 33:2169-2185
Huang, S., K.A. Rahn, and R. Arimoto	Semiannual cycles of pollution at Bermuda	Journal of Geophysical Research, 104:30309-30318

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**Appendix D. Presentations and Publications during 1999 (Continued)**

<b>Author</b>	<b>Title</b>	<b>Publisher/Conference</b>
Kirchner, T.B.	Uncertainty analysis	Calculating and Understanding Risk from Chemicals Released to the Environment, San Antonio, Texas
Kirchner, T.B.	Introduction to uncertainty analysis	Calculating and Understanding Risk from Chemicals Released to the Environment, Seattle, Washington
Kramer, G.H., M.A. Lopez, and J. Webb	A joint HML-CIEMAT-CEMRC project: testing a function to fit counting efficiency of a lung counting germanium detector array to muscle equivalent chest wall thickness and photo energy using a realistic torso phantom	Radiation Protection Dosimetry, submitted
Malek, M., T.G. Hinton, and S.B. Webb	Comparative uptake pathways of <sup>137</sup> Cs and <sup>90</sup> Sr in cabbage grown near Chernobyl	Health Physics, in press
Maring, H., D.L. Savoie, M.A. Izaguirre, C. McCormick, R. Arimoto, J.M. Prospero, and C. Pilinis	Aerosol physical and optical properties and their relationship to aerosol composition in the free troposphere at Izana, Canary Island during July 1995	Journal of Geophysical Research, submitted
N/A	Online/In Print, feature item	Environmental Science & Technology 133(17):382A
Orcutt, K.M., F. Lipschultz, K. Gundersen, R. Arimoto, A.F. Michaels, A.H. Knap, and J.R. Gallon	Seasonal pattern and significance of N <sub>2</sub> fixation by <i>Trichodesmium</i> spp. At the Bermuda Atlantic Time-series Study (BATS) site	Deep Sea Research II Special Issue: Nitrogen Fixation by <i>Trichodesmium</i> in the Sargasso Sea, submitted
Usman, S., H. Spitz, L. Shoaib, and S.C. Lee	Analysis of electret ion chamber radon detector response to interference from ambient gamma radiation	Health Physics 76(1):44-49
Tomza, U., R. Arimoto, and B.J. Ray	Filter color as an indicator of aerosol composition	Atmospheric Environment, submitted
Webb, J. and G.H. Kramer	An evaluation of germanium detectors employed for the measurement of radionuclides deposited in lungs using an experimental and Monte Carlo approach	Health Physics, submitted
Webb, J., D. Schoep, R. Arimoto, T. Kirchner, B. Stewart, and S. Webb	Environmental monitoring of radioactive and elemental constituents released as aerosols from the DOE WIPP	45 <sup>th</sup> Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry, Gaithersburg, Maryland

Table continued on next page

### Appendix D. Presentations and Publications during 1999 (Continued)

Author	Title	Publisher/Conference
Webb, J., D. Schoep, R. Arimoto, T. Kirchner, B. Stewart, and S. Webb	Environmental monitoring of radioactive and elemental constituents released as aerosols from the DOE WIPP	Workshop on WIPP Radiation Air Monitoring Programs, Carlsbad, New Mexico
Webb, J.	An evaluation of 80-mm diameter, 20-mm thick, broad energy germanium detectors for the measure of radionuclides deposited in lungs	45 <sup>th</sup> Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry, Gaithersburg, Maryland
Webb, J.	An overview of the CEMRC in vivo bioassay and quality control programs	45 <sup>th</sup> Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry, Gaithersburg, Maryland
Webb, J., M. Gadd, F. Bronsen and O. Tench	An evaluation of recent technology, including broad energy germanium detectors (BEGe) employed by Canberra Industries to improve measurement sensitivities of radionuclides deposited in lungs	In Vivo 99, Mol, Belgium
Webb, J. and T. Kirchner	<i>In vivo</i> measurements sensitivity and occurrence of radionuclides in residents of the Carlsbad, New Mexico area	In Vivo 99, Mol, Belgium
Webb, J. and T. Kirchner	<i>In vivo</i> measurements of residents in the Carlsbad, New Mexico area	Radiation Protection Dosimetry, submitted
Webb, J., M. Gadd, F. Bronsen and O. Tench	An evaluation of recent lung counting technology	Radiation Protection Dosimetry, submitted
Webb, J.	The cow counter: technology for the measure of radio-contaminants and fat-free lean content in livestock	WERC 1999 Conference on the Environment, Albuquerque, New Mexico
Webb, J.	Overview of the Carlsbad Environmental Monitoring & Research Center	WERC 1999 Conference on the Environment, Albuquerque, New Mexico
Webb, S., S.A. Ibrahim, F.W. Whicker	Inventory estimate of <sup>239</sup> Pu in soils east of Rocky Flats, Colorado	Environment International, in press
Zhang, X.Y., R. Arimoto, Z. S. An, J. J. Cao and D. Wang	Atmospheric dust aerosol over the Tibetan Plateau	Journal of Geophysical Research-Atmospheres, submitted
Zhang, X.Y., R. Arimoto, G.H. Zhu, T. Chen, and G.Y. Zhang	Concentration, size-distribution and deposition of mineral aerosol over Chinese desert regions	Tellus 50B:317-330
Zhang, X.Y., R. Arimoto, and Z.S. An	Glacial and interglacial patterns for Asian dust transport	Quaternary Science Reviews 18:811-820

### Appendix E. Guest Colloquia

Topic	Group/Event
Detecting neutrinos from supernovae: why and how?	Richard Boyd, Professor, Department of Physics, Ohio State University
Assessment of plutonium and americium in wounds	Milan Gadd, Senior Health Physicist, Rocky Flats Environmental Technology Site
Assessing and monitoring rangeland health	Walter Whitford, Senior Research Ecologist, U.S. Environmental Protection Agency
Environmental radioactivity: the interface between science and models	Ward Whicker, Professor, Department of Radiological Health Sciences, Colorado State University
Corrosion residues and punk rock from Lechuguilla Cave	Penelope Boston, Director of Research, Complex Systems Research, Inc. and Michael Spilde, Manager, Scanning Electrode Laboratory, University of New Mexico
Actinide analyses by thermal ionization mass spectrometry	Ross Williams, Manager (former), Radiological Services, American Environmental Network, Inc.
Trace elements in the atmosphere over the North Atlantic	Suilou Huang, Postdoctoral Fellow, New Mexico Institute of Mining and Technology
Transport of hydrophobic organic chemicals in aquatic systems	Richard Jepsen, Senior Technical Staff, Sandia National Laboratories
Applications of short-lived radionuclides as tracers and chronometers in earth science: an overview	Mark Baskaran, Department of Marine Sciences, Texas A & M University at Galveston
Resuspension studies at the WIPP site	Jeff Whicker, Health Physics Measurement Group, Los Alamos National Laboratory
Diversity and life history variation among ground-dwelling sheet-web spiders	Michael Draney, Postdoctoral Fellow, Department of Biology, New Mexico State University
Geochemical performance of German low level waste disposal facilities	Bernhard Kienzler, Institut fuer Nukleare Entsorgungstechnik, Germany
The inventory and distribution of $^{129}\text{I}$ in sagebrush steppe at the Idaho National Engineering and Environmental Laboratory	Randall Morris, Research Scientist, Environmental Science & Research Foundation
The role of colloid dispersion in the remediation of abandoned oil field brine pits	Mark Walthall, Associate Professor, Agronomy Department, Louisiana State University
Selected approaches to evaluating the fate and effects of contaminants in freshwater systems	Kurt Maier, Assistant Professor, Department of Biology, University of Memphis
NIST traceability – how good is good enough?	Kenneth Inn, Group Leader, Office of Radiation Measurements, National Institute of Standards and Technology
Altering coastal plain landscapes for fun (quail hunting) and profit (agriculture)	William Michener, Associate Scientist, J.W. Jones Ecological Research Center
Aerosols, optics and aesthetics: the battle to save Lake Tahoe	Thomas Cahill, Professor Emeritus, Department of Atmospheric Sciences, University of California, Davis



**Appendix F. Major Tours, Public Presentations and Exhibits**

Group/Event
Environmental Management Seminar, NMSU – two guest lectures
Biology classes from NMSU-Carlsbad – presentation & tour
Representatives for New Mexico Business Journal – tour
Geology students, Iowa State University – tour and presentation
Chihuahuan Desert Conservation Alliance – tour and presentation
Chihuahuan Desert Laboratory class from NMSU-Carlsbad – tour and presentation
Opening Ceremonies for the WIPP - exhibit
Presentation on CEMRC and local water quality – Project WET workshop for Carlsbad teachers
1999 SCERP Border Environmental Technical Exchange and Conference - exhibit
Student science day sponsored by AAUW – tour and presentation
Living Desert State Park staff - presentation
Project Uplift student group – tour and presentation
Carlsbad Ham Radio Club – presentation
International Good Neighbor Council – presentation
Carlsbad Board of Realtors - presentation
Carlsbad Public Schools Science Showcase – exhibit “What Can You Tell about the Weather?”
Carlsbad Lions Club - presentation
Carlsbad Art Association - presentation
Department of Geosciences, Texas Tech University – two guest lectures
American Association of University Women – presentation and tour
Carlsbad Municipal Schools, P.R. Leyva Middle School, 6 <sup>th</sup> grade classes – tour and program

**Appendix G. Leadership Participation by CEMRC Staff  
in Professional Functions**

Function	CEMRC Staff/Role
ACE-Asia	R. Arimoto, Member, Executive Committee
International Global Atmospheric Chemistry/Asia Pacific Regional Experiment, Seoul, Korea	R. Arimoto, Member, Executive Committee
American Geophysical Union, Journal of Geophysical Research-Atmospheres	R. Arimoto, Associate Editor
Loessfest '99, Bonn, Germany	R. Arimoto, Session Paper Reviewer
American National Standards Institute, HPS N13.25, Internal Dosimetry Programs for Plutonium Exposure – Minimum Requirements	J. Webb, Member, Standards Committee Working Group
45 <sup>th</sup> Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry, Gaithersburg, Maryland	J. Webb, Session Chair
In Vivo 99, Mol, Belgium	J. Webb, Session Paper Reviewer
45 <sup>th</sup> Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry, Gaithersburg, Maryland	B. Stewart, Presentor, Workshop “Radiochemistry 101”

### 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 D. Davis, Georgia Institute of Technology, and others)	National Science Foundation	\$160,000 (1998-2002)	Funded, in progress
Mineral dust and radionuclides over the North Atlantic	R. Arimoto (with R.A. Duce, Texas A&M University)	National Science Foundation	\$270,428 (1997-2000)	Amended, in progress
Characterization of ambient particulate matter in the Paso del Norte region	R. Arimoto (with five NMSU investigators)	U.S. Environmental Protection Agency, via Southwest Center for Environmental Research and Policy	\$27,843 (1999-2000)	Funded, in progress
Determination of Be and U in aqueous extracts of contaminated soils	R. Arimoto (with R.A. Cole and C.A Caldwell, NMSU)	U.S. Fish & Wildlife Service	\$5,618 (1999-2000)	Funded, in progress
Ambient air quality issues related to confined animal operations	R. Arimoto (with R.A. Duce, Texas A&M University, B. Auvermann, Texas Agricultural Extension Service, and J.L. Botsford, NMSU)	U.S. Department of Agriculture, National Research Initiative Competitive Grants Program	\$49,976 (1999-2001)	Funded, in progress
Science and implementation plan for the ACE-Asia Network studies	R. Arimoto (with M. Uematsu and the APARE Coordination Committee)	National Science Foundation	No request for funding	Submitted
Environmental analytical laboratory services for brine inorganics	R. Arimoto	Westinghouse Environmental Services Company	\$620,892 (1999-2005)	Submitted, not funded
Proposal to establish the U.S. Department of Energy Waste Isolation Pilot Plant Environmental Research Park	M. Conley	DOE/Carlsbad Area Office	No request for funding	Submitted
Long-term risk from actinides in the environment: modes of mobility	T. Kirchner (with D. Breshears, Los Alamos National Laboratory, and S.A. Ibrahim, Colorado State University)	DOE Office of Environmental Management	\$89,900 (1997-2000)	Funded, in progress

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**Appendix H. New Project Development (Continued)**

<b>Proposal/Bid Title</b>	<b>PI(s)</b>	<b>Sponsor</b>	<b>Funding (Term)</b>	<b>Status</b>
A component oriented system for building structurally adaptive ecological models	T. Kirchner	U.S. Environmental Protection Agency, STAR Program	\$371,906 (1999-2003)	Submitted, not funded
Tools and methodologies for managing uncertainties and improving accuracy in risk assessment models	T. Kirchner	National Science Foundation	\$422,912 (1999-2002)	Submitted, not funded
Limnological monitoring: Brantley Dam Reservoir	D. Schoep	U.S. Department of Interior, Bureau of Reclamation	\$56,379 (1997-2000)	Funded, in progress
Lung & whole body <i>in vivo</i> radiobioassay measurements	J. Webb	Waste Control Specialists, Inc.	\$233,414 (1997-2000)	Amended, in progress
<i>In vivo</i> radiobioassay measurements for WIPP personnel	J. Webb	Westinghouse Electric Company	\$573,000 (1998-2001)	Amended, in progress
<sup>210</sup> Pb - A biomarker for exposure of people to radon in indoor environments	J. Webb	Lovelace Respiratory Research Institute	\$116,182 (1999-2002)	Submitted, not funded
Internal dose assessments from historical radiation worker records	J. Webb	MJW Corporation	\$10,000 (1999)	Funded, in progress
Measurement of <sup>210</sup> Pb and <sup>7</sup> Be in environmental media using contrasting geometries	J. Webb	International Atomic Energy Agency	No funding requested	Submitted
The cow counter: technology for the measure of radio-contaminants and fat-free lean content in livestock	J. Webb (with G. Duff, NMSU)	DOE, via Waste-management Education and Research Consortium	\$169,860 (1999-2001)	Funded, in progress

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## **Appendix I. Status of Completion of 1999 Key Performance Indicators**

1. Concurrent high-volume and low-volume aerosol sampling at three locations through 1999. [Completed]
2. Collection of daily FAS samples in WIPP exhaust shaft through 1999. [Completed]
3. Collection of triplicate soil samples at current 32 locations during January-February 1999. [Delayed, completed March 1999]
4. Concurrent operation of meteorological sampling stations at two sites through 1999. [Completed]
5. Collect drinking water samples at 6 sources during March-April 1999. [delayed, completed August 1999]
6. Collect sediment and surface water samples from three reservoirs during June-July 1999. [delayed, completed August 1999]
7. Collect vegetation samples from six locations during spring and fall 1999. [No spring collection due to absence of new growth; fall collection completed]
8. Collect composite arthropod sample from one location during 1999. [Completed; collection expanded to two locations]
9. Continue *in vivo* bioassays for public. [Bioassays for over 350 people completed prior to waste receipt at WIPP, initial measurements for over 40 new volunteers; repeat measurements initiated in July 1999.]
10. Radioanalyses of all 1998 aerosol, soil, sediment, surface water, drinking water and vegetation samples by October 1999. [Not completed; radioanalyses completed for 1998 soils, sediment, surface water and drinking water]
11. Radioanalyses of FAS sample analyses to meet quarterly posting schedule. [Completed for gross alpha, gross beta and gamma results; delayed for actinides, completed September]
12. Inorganic trace element analyses of representative subset of low-volume aerosol, soil, sediment, surface water and drinking water samples within three months after each sample collection. [Completed]
13. Implement electronic Laboratory Information Management System by October 1999. [Completed for FAS data]
14. Post results of radioanalyses of 1998 samples within two months after completion of analyses of each set of samples. [Completed for soils, FAS, drinking water and surface water samples; delayed for all other media.]
15. Post results of non-radiological analyses of 1999 samples within two months after completion of each set of samples. [Completed for FAS samples, delayed for all other media]
16. Make CEMRC 1998 Report and background data accessible via Internet by March 1999. [Completed]
17. Submit manuscript for publication by July 1999 on radioanalyses of soils. [Delayed, scheduled for February 2000]

## **Appendix J. CEMRC Quality Assurance Policy**

The Carlsbad Environmental Monitoring & Research Center (Center) is a division of the College of Engineering, New Mexico State University (NMSU). The Center 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. In addition to the general goals, mission and standards of NMSU, the Center adheres to the following principles:

- Standards of quality assurance and quality control incorporating standard scientific methods will be developed and implemented that are appropriate to the objectives and functions of specific projects and programmatic areas.
- Methods for performance assessment and quality improvement will be used throughout the Center in keeping with policies and procedures of NMSU, and with protocols adopted for specific projects and programmatic areas.
- Personnel, equipment and facilities will be provided to achieve adopted project objectives and quality standards, subject to the limitations of fiscal and other applicable constraints.
- Personnel will be provided access to written and verbal guidance, training and other professional development to support continuous improvement within all programmatic areas, subject to the limitations of fiscal and other applicable constraints.
- Personnel will be held accountable for their actions related to protection of employees, the public and the environment, in carrying out projects and other activities, in compliance with all applicable laws and regulations.
- Employees are responsible for personal compliance with policies, procedures and other guidance adopted for purposes of quality control, fiscal accounting and other management objectives.

## **Appendix K. Quality Assurance/Quality Control for 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, 1983, *Methods for Chemical Analysis of Water and Wastes*, EPA/600/4-79-020; EPA, 1997, *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*, EPA/SW-846; American Public Health Association, 1981, *Standard Methods for the Examination of Water and Wastewater, 20<sup>th</sup> Edition*). 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.

### **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 aspiration through an air/acetylene flame, by vaporization in a heated graphite furnace, by flow-injection via a heated quartz cell, or through an unheated quartz cell (for Hg). Additional inorganic analyses were performed using a Perkin-Elmer Optima 3300 dual-view, inductively-coupled plasma atomic (or optical) emission spectrometer (ICP-ES). In February 1999, the Optima ICP-ES was replaced by a Perkin-Elmer Elan 6000 inductively-coupled plasma mass spectrometer (ICP-MS). The three instruments used for the elemental analyses are complementary; AAS is more sensitive than the ICP-ES and the ICP-MS for some elements, especially for the hydride elements (As, Se and Hg), but compared with the ICP-ES and 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. The ICP-MS is much more sensitive than the ICP-ES, lowering detection limits by ten to a hundred times for some analytes, and allowing analyses for substantially more elements, including the rare earths.

### **General Quality Control**

Several analytes are readily determined by more than one of the four instruments used at CEMRC, which facilitates intra-laboratory comparisons as summarized below. Some of these internal QC comparisons are also summarized in the sections of this report that deal with specific media.

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 value 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 quality control verifications and batch QC provide records of sample performance data. Copies of the analytical data and performance results are maintained in the environmental chemistry instrument laboratory. The laboratory also carried out several informal inter-laboratory comparisons, and has recently begun participation in performance testing under the National Voluntary Laboratory Accreditation Program (NVLAP); no NVLAP results had been received by the end of 1999.

Calibrations are verified with a standard obtained from a source different from that used in procuring the primary calibration standards. The calibration standards and the verification standards

used at CEMRC are, where possible, traceable to NIST. A calibration blank is analyzed at the beginning of each workday when samples will be run, after every ten samples, and at the end of the day. To pass the calibration verification, blank results must be less than the minimum detectable level or  $\pm 3$  SD of control limits. Analysis of a blank and a standard are performed at a frequency of 10% during analytical runs, and these are repeated at the close of each analytical run to verify continued calibration validity. Batch quality control samples are counted as samples in determining the 10% frequency, but the continuing check samples are not counted as samples in determining the 10% frequency.

Various types of field blanks, check solutions and laboratory fortified (spiked) samples are analyzed along with the samples as part of the QA/QC procedures. These vary somewhat among matrices and analyses as described in more detail below. In addition, when feasible, duplicate samples are processed to evaluate reproducibility and sample homogeneity. Control charts for each matrix have been established, and  $\pm 3$  SD limits have been determined for future reference. Control charts are used to track the performance of the instrument and the sample preparation procedures. Similarly, spike recoveries are calculated, tracked and reported along with the analytical data.

### **Quality Control for Analyses by IC**

For the IC 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.

Method Detection Limits (MDL) were established for each analyte in each sample matrix according to EPA Method 300.0 (Determination of Inorganic Anions by Ion Chromatography) (Table K1). Fluoride was not determined in aerosol filters and soils due to co-eluting organic peaks, but method development is underway to correct this. QC samples included Laboratory Reagent Blanks (LRB), with one LRB prepared for each sample batch (normally a set of ten samples). LRB results below MDL are considered acceptable (Table K2). For aerosol filter analyses, some LRB results indicated reagent blank contamination, and this was subsequently identified and eliminated; the samples could not be reanalyzed because the filters are consumed in the analysis process. Laboratory Fortified Matrix (LFM) samples were also used for QC, with one LFM analysis per sample batch. Results from analyses of LFMs are used to calculate matrix spike recoveries, with recoveries of 70-130% considered acceptable. As prescribed by EPA Method 300.0, chloride and sulfate values in water samples and chloride and sulfate values in sediments were not reported because the concentration of the fortification was less than 25% of the background concentration (Table K3).

One duplicate analysis was performed for each sample batch. When feasible, duplicate aliquots of some field samples were analyzed. In cases where duplicate aliquots from the original sample were not feasible (such as aerosol filters), separate aliquots of the sample extract were analyzed. The relative percent difference (RPD) between the sample and the duplicate was calculated, with a difference of  $< 20\%$  (or an absolute difference of  $\pm$  MDL for samples less than five times the MDL) considered acceptable (Table K4). Sulfate duplicates in water and sulfate and chloride in sediments were not within limits because the sample concentrations were beyond the instrument's calibration of 50 ppm for chloride and 100 ppm for sulfate. The duplicates were not analyzed at the diluted level.

A Laboratory Fortified Blank (LFB) was prepared and analyzed with each sample batch, using a spiked ultrapure water sample for aerosol filters and water samples, and certified reference materials (CRM) for soils and sediments. Recoveries of 85-115% were considered acceptable for aerosol filters and water samples, and the analyses of the LFBs produced values well within this range (Table K5). The CRM was "Anions in Soils" from Environmental Research Associates (ERA) in Arvada, Colorado and it was used as the reference material for both soils and sediments. Use of an end-over-end rotator was necessary to achieve the limits set by ERA for this CRM (Table K6). Because there is no existing standard reference method for extracting soils or sediments for anion analysis, the results obtained by different methods may not be directly comparable.

Low-volume aerosol filters were also analyzed by IC for five cations with overall acceptable results (Table K7). Acceptance limits for each QC parameter were the same as previously described.

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## Quality Control for Elemental Analyses by ICP-ES, ICP-MS and AAS

For elemental analyses, sets of quality control samples comparable to those previously described for the IC analyses were included with each sample batch. Detailed performance results for all QC measures are not presented here due to the number of elements that are determined by ICP-ES, ICP-MS and AAS. For all media (aerosol filters, water, soils and sediments), ICP-ES, ICP-MS and AAS values were reported relative to the method detection limit as determined by EPA protocols (Table K8 and K9). Digestion QC samples were analyzed at a frequency of 10% relative to samples. The digestion QC control parameters used for the evaluation of metals in aerosol filters included LRB filters and vendor-supplied certified reference filters. Due to sample volume limitations, duplicate and post digestion spike analyses could not be performed for the ICP-ES or ICP-MS analyses of the aerosol samples.

For water, soils and sediments, a practical quantitation limit (PQL) was also calculated to evaluate precision based on the analysis of duplicate samples. The PQL is obtained by multiplying the method detection limit (MDL) by five. The digestion quality control parameters used for the evaluation of metals in water, soils and sediments were based on EPA Contract Laboratory Program (1994, U.S. EPA Contract Laboratory Program National Functional Guidelines for Inorganic Data Review, EPA 540/R-94013) and SW846 methods (EPA, 1997, *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*, EPA/SW-846). No comparable control parameters presently exist for aerosol samples.

For aerosol samples, unused cellulose ester filters were used as LRB samples. LRB results above the MDL were subtracted from each associated batch of sample results, because the LRB results were greater than the MDL for many of the analytes studied. Analysis of reagent digests have shown the contamination to be inherent to the cellulose ester filters for some of these analytes (Ca, Cr, Cu, Mg, Ni and Pb), while others (Ce and Sm) are introduced in trace amounts by the reagents used for digestion. A cellulose ester CRM ("Trace Metals on Filter Media" from High Purity Standards in Charleston, South Carolina) was also used for QC of aerosol sample analysis. Mean recoveries for all analytes were within 15% of the manufacturer's established true values.

For FAS samples, unused Versapore® filters were used as LRB samples. LRB results above the MDL were subtracted from each associated batch of sample results, because the LRB results were greater than the MDL for several of the analytes studied. Analysis of reagent digests have shown the contamination to be inherent in the Versapore filters for several analytes (Cu, K, La, Mg, Na, Nd, Ni, Pb and Sm), while others (Dy, Gd) are introduced in trace amounts by the reagents during digestion. A cellulose ester CRM ("Trace Metals on Filter Media" from High Purity Standards in Charleston, South Carolina) was used for QC of the FAS samples. Mean recoveries for all analytes were within 15% of the manufacturer's established true values, with the exception of Cu. The filter fortified by High Purity Standards has a much lower Cu background than the Versapore filter and the Cu level contained in the Versapore filters is significant compared to the fortification level, producing a low bias in CRM recovery when blank subtraction is used.

Four standard QC measures were used in association with analyses of drinking water and surface water samples. Ultrapure water was used for LRB samples and average concentrations were less than the MDLs for all analytes except Tl, Th and Ca in drinking water samples and As, Cr, Ti, Ni, Sr, Ag, Sb, Ba, Th, U, Na and Ca in surface water samples. The amounts of Sr, Ba, U, Na and Ca in the samples were so much higher than in the reagent blanks that the contribution from the blank was negligible. The sample values for Tl and Th in drinking water may be biased 21% and 12% high, respectively, because blank subtraction was not performed. Likewise, surface water results for Cr, Ti, Ni, Sb and Th are biased 67%, 0.2%, 25%, 122% and 40% high. Surface water results for Ag were all less than the MDL. Samples were not blank corrected following the procedure prescribed by the EPA Contract Laboratory Program (1994, U.S. EPA Contract Laboratory Program National Functional Guidelines for Inorganic Data Review, EPA 540/R-94013). However, the sample values for As were corrected for the reagent blank recoveries to correct for the slight high bias (10%). A LFB was prepared by adding a known quantity of each analyte of interest to ultrapure water. All analytes for

drinking water and surface water had recoveries within the 85-115% limits as specified by EPA methods.

LFM samples were also used for QC in analyses of water samples, with all recoveries within the 70%-130% acceptance window, with the exception of Sr (0%) in drinking water and in surface waters (0%). If the concentration of the fortification is less than 25% of the background concentration, the recovery of the LFM is usually not reported. In this case, the background concentration of Sr in drinking water was 1680%, and Sr in surface water was 455%, so the results were within acceptance limits. A duplicate digestion analysis of water samples was also performed to demonstrate reproducibility, but a slight modification of the EPA CLP program was used for acceptance determination. If the sample result was less than the PQL, a  $\pm$  PQL control limit was used. If the sample result was greater than the PQL, a  $\pm 20\%$  RPD control limit was used. All duplicate results were within these modified acceptance limits with the exception of Cr, Ag, Sb and Th in drinking water and Gd, Th, As and Se in surface water. In compliance with the EPA Contract Laboratory Program (1994, U.S. EPA Contract Laboratory Program National Functional Guidelines for Inorganic Data Review, EPA 540/R-94013), the results for these analytes should be considered estimates.

For soils and sediments, LRB samples of ultrapure water were compared to MDLs to determine if contamination was introduced during sample preparation. LRB results were within acceptance limits for soils with the exception of Be, Ni, Ag, Sb, Ce, Pr, Nd, Pr, Nd, Th and Ca, which were above the MDL. However, the sample measurements were at least ten times higher than the LRB results for Be, Ce, Pr, Nd, Th and Ca and therefore the contaminant effects on the measurements were considered negligible. Sample results were not corrected by blank subtractions, therefore results may be biased high for Ni, Ag and Sb. Sediment LRB results were less than the MDL with the exception of the following analytes: Be, Ni, Zn, Sr, Sb, La, Eu, Th, U and Ca. The elemental concentrations of all analytes, excluding Sb, in sediment samples were several orders of magnitude higher than LRB results, and therefore the contaminant effects on measurements in sediments are considered negligible. Sb results in sediments may be biased high because the results were not corrected for the laboratory reagent blank.

A CRM ("Priority Pollutant T/CLP Soil" from ERA) was obtained and prepared with the soil and sediment samples to demonstrate matrix-specific performance of digestion and analysis procedures. All analytes except Se were recovered within the supplier's specified control limits for all digestions. The Se CRM results recovered slightly high for two of the soil batches. However, the Se concentrations in all samples associated with these batches with high CRM recoveries were at or below the MDL, therefore re-digestion for Se was not performed. Duplicate digestions were performed for soil and sediment using a modification of the EPA CLP program for acceptance determination. If the sample result was less than the PQL, a  $\pm$  PQL control limit was used. If the sample result was greater than the PQL a  $\pm 20\%$  RPD control limit was used. For soils and sediments, the average RPD over the digestions performed was within acceptance limits for all analytes, except Hg, Se and Tl and the absolute difference on the duplicates was used to determine that these analytes were still within limits. A LFM also was prepared with an average recovery within 70%-130% windows for all analytes with the exception of Sb at 33%. Mn, Ba, Sb and Sr in sediments recovered at 0%, 0%, 37% and 774%, respectively. A low bias for Sb was expected due to the digestion procedure used, as noted in the CEMRC 1998 Report. For Mn, Ba and Sr in sediment, the concentration of the fortification was less than 25% of the background concentration (Mn 106%, Ba 103%, Sr 109%), so the results for these analytes were acceptable.

## Conclusions and Future Improvements

In IC analyses, development is in progress to improve the resolution of fluoride and its separation from co-eluting organic species. As a result of this effort, quantification of acetate and formate should be possible for certain types of samples. Although CEMRC has already demonstrated low elemental MDLs, these could be improved for some analytes (including Sn, Zn, Na, Mg, Al, K, Ca, Ni, Pb, Mn and Fe) by reducing reagent blank contamination from the acids used to prepare standards and samples. In addition to the double distillation already in use, elimination of metal corrosion in-

side fume hoods is being undertaken to further reduce trace contamination. Another method is also being developed to prepare soils and sediments via closed vessel microwave digestion, which should also improve MDLs and increase sample throughput. As noted in the matrix-specific descriptions, blank-correction for results was inconsistently applied among media types. A blank-correction standard is under development for adoption in future analyses.

**Table K1. Method Detection Limits for Analyses by IC**

Sample Matrix	Unit Type (Units)	Fluoride	Chloride	Nitrite	Nitrate	Phosphate	Sulfate
Low volume aerosol filter (AS11 column)	General (ug L <sup>-1</sup> )	<sup>a</sup> NA	34.8	101	22.9	8.54	16.7
Low volume aerosol filter (AS14 column)	General (ug L <sup>-1</sup> )	NA	25.3	NA	25.3	48.5	11.1
Drinking water and surface water	General (ug L <sup>-1</sup> )	48.7	21	NA	58.8	235	111
Soil	General (ug L <sup>-1</sup> )	NA	34.8	101	22.9	8.54	16.7
Sediment	General (ug L <sup>-1</sup> )	48.7	21	NA	58.78	235	111
<sup>b</sup> Low volume aerosol filter (AS11 column)	Matrix specific (ug m <sup>-3</sup> )	NA	0.0372	0.1081	0.0045	0.0245	0.0179
<sup>b</sup> Low volume aerosol filter (AS14 column)	Matrix specific (ug m <sup>-3</sup> )	NA	0.0271	NA	0.0271	0.0519	0.0119
<sup>c</sup> Drinking water and surface water	Matrix specific (ug m <sup>-3</sup> )	48.68	20.96	NA	58.75	235.3	110.7
<sup>d</sup> Soil	Matrix specific (mg kg <sup>-1</sup> )	NA	0.5604	0.0844	0.2386	0.2793	0.1496
<sup>d</sup> Sediment	Matrix specific (mg kg <sup>-1</sup> )	0.4868	0.2096	NA	0.5875	2.3531	1.107

<sup>a</sup>NA = not analyzed

<sup>b</sup>Teflo® 0.2 micron 45 mm diameter filter extracted into 30 mL ultrapure water; nominal flow volume of 28 L<sup>3</sup> of air per filter

<sup>b</sup>Water samples are analyzed by direct injection

<sup>c</sup>5 g of solid material extracted into 50 mL ultrapure water

**Table K2. Mean Laboratory Reagent Blank Results for IC**

Sample Matrix	Units	Fluoride	Chloride	Nitrite	Nitrate	Phosphate	Sulfate
Low volume aerosol filter (AS11 column)	ug L <sup>-1</sup>	<sup>a</sup> NA	9.61	34.28	22.87	8.54	16.73
Low volume aerosol filter (AS14 column)	ug L <sup>-1</sup>	NA	11.8	NA	14.4	0	1.8
Drinking water and surface water	mg L <sup>-1</sup>	0	0	0	0	0	0
Soil	mg kg <sup>-1</sup>	NA	.0886	.0032	.1937	.0736	.1452
Sediment	mg kg <sup>-1</sup>	0	0	0	0	0	0

<sup>a</sup>NA = not analyzed

**Table K3. Mean Laboratory Fortified Matrix Recovery Results for IC**

Sample Matrix	Fluoride	Chloride	Nitrate	Phosphate	Sulfate	% Recovery Limit
Low volume aerosol filter	<sup>a</sup> NA	104%	107%	103%	100%	70-130%
Drinking water/surface water	87%	<sup>b</sup> NR	95%	63%	NR	70-130%
Soil	NA	107%	103%	101%	98%	70-130%
Sediment	91%	NR	95%	74%	NR	70-130%

<sup>a</sup>NA = not analyzed

<sup>b</sup>NR = not reported

**Table K4. Mean Relative Percent Difference Results for Ion Chromatography**

Sample Matrix	Fluoride	Chloride	Nitrite	Nitrate	Phosphate	Sulfate	% Recovery Limit
Low volume aerosol filter	<sup>a</sup> NA	-1%	0	-1%	-12%	0%	+/- 20
Drinking water, surface water	0.18%	4.58%	NA	-3.3%	0%	26.73%	+/- 20
Soil	NA	5.12%	NA	0.59%	1.21%	0.75%	+/- 20
Sediment	0%	77%	NA	0%	0%	-99%	+/- 20

<sup>a</sup>NA = not analyzed

**Table K5. Mean Laboratory Fortified Blank Recovery Results for Ion Chromatography**

Sample Matrix	Fluoride	Chloride	Nitrate	Phosphate	Sulfate	% Recovery Limit
Low volume aerosol filter	<sup>a</sup> NA	107%	101%	102%	99%	85-115%
Drinking water and surface water	94%	102%	96%	91%	95%	85-115%

<sup>a</sup>NA = not analyzed

**Table K6. Mean Results for Standard Reference Materials using Ion Chromatography**

<sup>a</sup> Sample Matrix	Analyte (mg kg <sup>-1</sup> )				
	Fluoride	Chloride	Nitrate as N	Phosphate as P	Sulfate
Soil	<sup>b</sup> NA	17.37	38.86	16.48	25.04
Sediment	NA	15.67	36.39	19.26	24.59
Lower limit	NA	13.68	37.06	12.66	22.52
Upper limit	NA	18.52	50.14	17.13	30.47

<sup>a</sup>Standard reference materials (“Anions in Soil”) were prepared using soil procedures and sediment procedures separately; lower and upper limits are vendor-provided values

<sup>b</sup>NA = not analyzed

**Table K7. QC Results for Cations in Aerosol Filters Analyzed by Ion Chromatography**

Parameter (units)	Analyte				
	Sodium	Ammonium	Potassium	Magnesium	Calcium
<sup>a</sup> MDL (µg L <sup>-1</sup> )	97.3	43.9	56.9	20.9	68.0
MDL (µg m <sup>-3</sup> )	0.104	0.047	0.061	0.022	0.073
Average <sup>b</sup> LRB (µg L <sup>-1</sup> )	5.4	19.5	-0.002	-0.25	-22.0
Average <sup>c</sup> LFM (%)	103%	109%	103%	104%	105%
Average <sup>d</sup> RPD (%)	17%	-9%	5%	5%	-7%
Average <sup>e</sup> LFB (%)	101%	96%	100%	97%	102%

<sup>a</sup>MDL = method detection limit

<sup>b</sup>LRB = laboratory reagent blank

<sup>c</sup>LFM = laboratory fortified matrix

<sup>d</sup>RPD = relative percent difference

<sup>e</sup>LFB = laboratory fortified blank

**Table K8. Method Detection Limits for Analyses by ICP-ES and AAS**

Instrument	Analyte	Units	Method Detection Limit			Accuracy	Precision
			<sup>a</sup> Air Filter	Water	Soil: Acid Extract		
ICP-ES	Al	ppm	0.0114	0.0227	0.242	± 30%	± 10%
ICP-ES	Sb	ppm	0.0026	0.0046	0.9172	± 30%	± 10%
	Ba	ppm	0.0002	0.0001	0.13	± 30%	± 10%
	Be	ppm	0.0002	0.0001	0.0156	± 30%	± 10%
	Bi	ppm	<sup>b</sup> NA	0.0031	0.6288	± 30%	± 10%
	Cd	ppm	0.0006	0.0003	0.0002	± 30%	± 10%
	Ca	ppm	0.0044	0.1189	2.08	± 30%	± 10%
	Cr	ppm	0.0006	0.0008	0.1658	± 30%	± 10%
	Co	ppm	0.0007	0.0002	0.0440	± 30%	± 10%
	Cu	ppm	0.0003	0.0010	0.1922	± 30%	± 10%
	Fe	ppm	0.0024	0.0164	1.42	± 30%	± 10%
	Pb	ppm	0.0026	0.0007	0.1473	± 30%	± 10%
	Mg	ppm	0.0020	0.0109	0.87	± 30%	± 10%
	Mn	ppm	0.0002	0.0001	0.0249	± 30%	± 10%
	Mo	ppm	0.0006	0.0002	0.031	± 30%	± 10%
	Ni	ppm	NA	0.0002	0.0347	± 30%	± 10%
	K	ppm	0.0153	0.0175	NA	± 30%	± 10%
	Na	ppm	NA	0.1332	0.145	± 30%	± 10%
	Sr	ppm	0.0006	0.0004	0.14	± 30%	± 10%
	V	ppm	0.0038	0.0007	0.1492	± 30%	± 10%
	Zn	ppm	0.0002	0.0025	0.24974	± 30%	± 10%
AAS ( <sup>c</sup> CV)	Hg	ppt	NA	6	6	± 30%	± 20%
AAS ( <sup>d</sup> FIH)	As	ppt	NA	30	150	± 30%	± 20%
	Se	ppt	NA	120	600	± 30%	± 20%
AAS ( <sup>e,f</sup> GF)	As	ppb	1.4	NA	1.4	± 30%	± 20%
	Se	ppb	1.2	NA	1.2	± 30%	± 20%
	Fe	ppb	29	11	NA	± 30%	± 20%

<sup>a</sup>µg per filter

<sup>b</sup>NA = not analyzed

<sup>c</sup>CV = Cold vapor

<sup>d</sup>FIH = Flow injection hydride

<sup>e</sup>GF = Graphite furnace

<sup>f</sup>Sb, Cd, Cr, Co, Cu, Pb, Ni and Zn can be analyzed by graphite furnace, but these analytes were determined by ICP-MS for this report.

**Table K9. Method Detection Limits for Analyses by ICP-MS**

Analyte	Method Detection Limit by Matrix (units)				Accuracy	Precision
	Air Filter ( $\mu\text{g filter}^{-1}$ )	Drinking Water ( $\mu\text{g L}^{-1}$ )	Surface Water ( $\mu\text{g L}^{-1}$ )	Soil/Sedimen t ( $\text{mg kg}^{-1}$ )		
Li	0.0215	0.1102	0.5304	0.0308	30%	20%
Be	0.0032	0.0359	0.0291	0.0015	30%	20%
V	0.0435	0.3652	0.5086	0.5486	30%	20%
Cr	0.40	0.1230	0.3847	0.3123	30%	20%
Ti	0.235	0.1188	0.3919	0.1466	30%	20%
Mn	0.0175	0.0086	0.5707	0.0880	30%	20%
Co	0.0145	0.0076	0.0339	0.0227	30%	20%
Ni	0.080	0.0217	1.7549	0.1260	30%	20%
Cu	0.0165	0.0886	1.6114	0.5789	30%	20%
Zn	0.650	0.2745	21.4771	2.7173	30%	20%
Sr	0.035	0.2098	0.5298	0.1191	30%	20%
Mo	0.036	0.0495	0.0241	0.0319	30%	20%
Ag	0.007	0.0018	0.0778	0.0049	30%	20%
Cd	0.003	0.0058	0.4628	0.0301	30%	20%
Sn	1.75	0.1211	19.5775	7.3785	30%	20%
Sb	0.0335	0.0056	0.2425	0.0169	30%	20%
Ba	0.0325	0.0068	0.4490	0.2233	30%	20%
La	0.0003	0.0004	0.0177	0.0017	30%	20%
Ce	0.0004	0.0007	0.0323	0.0032	30%	20%
Pr	0.0002	0.0007	0.0052	0.0007	30%	20%
Nd	0.0003	0.0023	0.0141	0.0014	30%	20%
Sm	0.0002	0.0030	0.0019	0.0007	30%	20%
Eu	0.0001	0.0010	0.0020	0.0005	30%	20%
Gd	0.0001	0.0021	0.0041	0.0007	30%	20%
Dy	0.0001	0.0017	0.0037	0.0006	30%	20%
Er	0.0002	0.0018	0.0014	0.0004	30%	20%
Tl	0.60	0.0384	7.9129	0.1365	30%	20%
Pb	0.0042	0.0430	1.9139	0.2244	30%	20%
Th	0.0002	0.0009	0.0056	0.0007	30%	20%
U	0.0002	0.0006	0.0019	0.0008	30%	20%
Na	7.55	3.7445	192.9923	58.0435	30%	20%
Mg	0.50	1.7066	18.7665	11.5821	30%	20%
Al	1.85	1.8070	17.3792	4.4980	30%	20%
K	2.65	12.9665	58.8624	39.1758	30%	20%
Ca	4.85	6.1419	146.4472	43.9966	30%	20%
Fe	<sup>a</sup> NA	1.0916	34.0169	11.8696	30%	20%
Hg	0.001	NA	NA	NA	30%	20%

<sup>a</sup>NA = not analyzed

## **Appendix L. Quality Assurance/Quality Control for Radioanalyses**

The CEMRC radioanalytical program conducted extensive method development throughout 1999, resulting in standard methodologies for determining background levels of alpha- and gamma-emitting radionuclides in soil, and for Pu in air filters. Methods were also developed for less sensitive measurements of alpha-emitting radionuclides in water and for gross alpha/beta and gamma measurements in aerosol samples from the WIPP exhaust airflow.

During 1999, the CEMRC radioanalytical program participated in two rounds of the National Institute of Standards and Technology Radiochemistry Intercomparison Program (NRIP). A Report of Traceability was received for measurements of two analytes in glass fiber filters and three analytes in water. The radioanalytical program also participated in the DOE Environmental Measurement Laboratory-Quality Assurance Program (EML-QAP), resulting in "acceptable" ratings for 45 individual determination of ten analytes in glass fiber filters, soil, vegetation and water samples (Table L1).

Daily (or each time the system is used) performance checks are done on the gas-flow,  $\alpha/\beta$  proportional counter used for the FAS program. These checks included counting  $^{239}\text{Pu}$  and  $^{90}\text{Sr}$  standards for efficiency control charting ( $2\sigma$  warning and  $3\sigma$  limits) and ensuring that  $\alpha/\beta$  cross-talk were within limits ( $\leq 10\%$  alpha into beta and  $\leq 0.1\%$  beta into alpha). Sixty-minute background counts are also recorded daily (count must be within the mean background  $\pm 3\sigma$ ). Standards made with  $^{152}\text{Eu}$  are counted daily on the coaxial and well, HPGe detector systems used for the drinking water, surface water, soil and FAS programs. Efficiency, centroid and resolution measurements were made and tracked using the detector system software (centroids within  $\pm 0.8$  keV and resolutions within the mean FWHM  $\pm 30\%$ ). Routine background determinations are made on the HPGe detector systems by counting blank samples and the data are used to blank correct the sample concentrations. Pulser checks were made on the alpha spectroscopy system each time a drinking water, surface water, soil or FAS sample was counted for actinides to ensure that peak centroid and resolution were acceptable (centroids within  $\pm 12$  keV and resolutions approved by the spectroscopist). Background counts and blanks are also routinely counted and tracked for the alpha spectroscopy system.

Other routine activities conducted for radioanalyses include (1) tracking and verification of analytical instrument performance, (2) use of ACS grade reagents, (3) use of ASTM Type II water for reagent preparations, (4) use of NIST traceable radionuclide solutions and (5) verification testing of radionuclide concentrations for tracers not purchased directly from NIST. In addition to analyte-surrogate isotopic tracers used in samples,  $^{148}\text{Gd}$  is added to samples where no alpha emitters are expected (e.g. thorium blanks) to provide a monitor of detector performance.

Blanks are used to identify contamination or interference carried through the actinide analytical process. Table L2 summarizes the results of reagent blank analyses completed while processing WIPP EM soil, water and FAS samples during 1999, which constituted approximately 10% of the sample load. Batches of samples where Pu or Am blanks were greater than MDC were reanalyzed. The results indicate that Pu and Am contamination (most likely from sample cross contamination) is detected, but infrequently, and the practice of analyzing blanks at least 10% of the time should be continued to monitor for contamination. Contamination of naturally occurring radioactive materials in blanks, especially  $^{228}\text{Th}$  and  $^{234}\text{U}$ , indicated that some reagents might contain trace quantities of these isotopes. An investigation to identify the source of the naturally occurring radionuclides is being conducted.

Isotopic tracers are used to determine the analytical system's effectiveness to extract, purify and quantify the isotopes of interest. Table L3 summarizes the tracer yields (recoveries), observed while processing WIPP EM soil, water, FAS and QA samples. Although some samples had tracer recoveries somewhat lower than the median values, they were adequate to meet minimum detectable concentration requirements. In general, the yields were greater for the EML-QAP, NRIP, water and FAS samples, which is likely because these samples were received and spiked as liquids, and the tracer (and analyte) were dissolved into the sample rather than being incorporated into a solid matrix as is done for WIPP EM soils. On the other hand, tracer recoveries were generally lower for blank sam-



ples, and this may indicate that the blank matrix behaved differently from the sample matrices in the radiochemical methods employed.

Analyses of replicate and split samples are used to estimate precision, which is analyte- and matrix-specific (Table L4). Approximately 10% of the sample load during WIPP EM soil analyses were split (duplicate aliquot) samples, and numerous replicates were also incorporated as part of the performance assessment activities. All results for replicate samples analyzed for EML-QAP met the ANSI 42.22 precision criterion of  $t < 3$  for differences between replicate samples. The relative percent difference (RPD) between isotope concentrations in WIPP EM split samples was greater for Pu and Am than for the naturally occurring radionuclides, which may result from their origin as fallout. The RPD was lower for EML-QAP replicate samples as compared to WIPP EM splits, again as would be expected for spiked, highly homogenized samples.

All WIPP EM split soil samples met the ANSI 42.22 standard for precision for  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$  and  $^{235}\text{U}$ . For other isotopes, 69-86% of the split samples met the ANSI standard; investigation of improvements for precision in these analytes is in progress. WIPP EM water and FAS samples were not split because of other QA requirements, e.g., MDC considerations.

**Table L1. Summary of Participation in Intercomparison Programs for Radioanalyses**

<sup>a</sup> Program	Media	Radionuclide	<sup>b</sup> Percent Bias	Number of Determinations	<sup>c</sup> Results
EML-QAP	Air Filter	<sup>241</sup> Am	7.5	1	Acceptable
		<sup>57</sup> Co	-6.0	1	Acceptable
		<sup>60</sup> Co	-0.8	1	Acceptable
		<sup>137</sup> Cs	-5.5	1	Acceptable
		<sup>238</sup> Pu	-2.2	1	Acceptable
		<sup>239</sup> Pu	0.0	1	Acceptable
		<sup>125</sup> Sb	18.1	1	Acceptable
		<sup>234</sup> U	8.3	1	Acceptable
		<sup>238</sup> U	6.6	1	Acceptable
	Soil	<sup>238</sup> Pu	-2.9	3	Acceptable
		<sup>239</sup> Pu	0.1	3	Acceptable
		<sup>234</sup> U	2.6	3	Acceptable
		<sup>238</sup> U	4.2	3	Acceptable
	Vegetation	<sup>241</sup> Am	-8.0	3	Acceptable
		<sup>244</sup> Cm	20.9	3	Acceptable
		<sup>238</sup> Pu	-6.4	3	Acceptable
		<sup>239</sup> Pu	4.2	3	Acceptable
	Water	<sup>241</sup> Am	6.4	2	Acceptable
		<sup>60</sup> Co	2.2	2	Acceptable
		<sup>137</sup> Cs	2.1	2	Acceptable
		<sup>238</sup> Pu	4.7	2	Acceptable
<sup>239</sup> Pu		6.0	2	Acceptable	
<sup>234</sup> U		10.0	2	Acceptable	
<sup>238</sup> U		11.6	2	Acceptable	
NRIP	Air Filter	<sup>241</sup> Am	-0.1	5	NIST Traceable, 5.9%
		<sup>238</sup> Pu	1.6	5	NIST Traceable, 6.4%
	Water	<sup>241</sup> Am	0.2	5	NIST Traceable, 2.0%
		<sup>238</sup> Pu	1.1	5	NIST Traceable, 2.0%
		<sup>238</sup> U	0.1	5	NIST Traceable, 1.9%

<sup>a</sup>EML-QAP = Environmental Monitoring Laboratory-Quality Assurance Program; NRIP = National Institute of Standards and Technology Radiochemistry Intercomparison Program

<sup>b</sup>Percent bias = difference between sponsor's known value for a sample, and the mean of measurements by CEMRC for the sample, expressed as a percent relative to the sponsor's value

<sup>c</sup>Results for EML-QAP or NRIP; "acceptable" is defined in Report EML-604, June 1999; NIST Traceable is defined under ANSI 42.22 standards at the stated limit; NR = results have not yet been reported by NIST

**Table L2. Laboratory Reagent Blank Results for Radioanalyses of Actinides in WIPP EM Samples**

Parameter	Analyte Activity Concentration								
	<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>241</sup> Am	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th
<b>Soil (mBq kg<sup>-1</sup>)</b>									
<sup>a</sup> N	15	15	12	14	14	14	14	14	14
<sup>b</sup> N>MDC	1	1	3	9	1	3	13	0	4
<sup>c</sup> Minimum	NA	NA	28.7	33.4	NA	25.9	105.8	NA	17.6
Median	62.8	65.9	31.2	47.6	15.5	35.2	151.6	NA	39.7
<sup>c</sup> Maximum	NA	NA	35.2	105.3	NA	77.9	3505	NA	82.1
<sup>d</sup> Ratio (%)	NA	NA	NA	0.6	3.8	0.4	1.6	NA	0.4
<b>Drinking Water and Surface Water (μBq L<sup>-1</sup>)</b>									
<sup>a</sup> N	3	3	3	3	3	3	3	3	3
<sup>b</sup> N>MDC	0	0	0	2	1	2	2	0	0
<sup>c</sup> Minimum	NA	NA	NA	369.3	NA	86.4	142.8	NA	NA
Median	NA	NA	NA	440.3	63.2	201.0	144.7	NA	NA
<sup>c</sup> Maximum	NA	NA	NA	511.3	NA	315.7	146.7	NA	NA
<b>FAS Quarterly Composite (μBq)</b>									
<sup>a</sup> N	2	2	2	2	2	2	2	2	2
<sup>b</sup> N>MDC	0	0	0	2	0	1	1	0	0
<sup>c</sup> Minimum	NA	NA	NA	6.2	NA	NA	NA	NA	NA
Median	NA	NA	NA	9.7	NA	9.6	8.1	NA	NA
<sup>c</sup> Maximum	NA	NA	NA	13.2	NA	NA	NA	NA	NA

<sup>a</sup>N = total number of samples

<sup>b</sup>N>MDC = number of samples with values greater than MDC (minimum detectable concentration)

<sup>c</sup>NA = not applicable for isotopes measured above MDC in <2 reagent blanks

<sup>d</sup>Ratio = ratio of median concentration of U and Th in blanks to the median U and Th isotope concentration in the 96 soil samples collected in 1998.

**Table L3. Laboratory Tracer Recovery Results for Radioanalyses of Actinides**

Analytical Group	Parameter	<sup>a</sup> Tracer Percent Recovery			
		Pu	Am	U	Th
<sup>b</sup> EML-QAP and NRIP	<sup>c</sup> N	14	15	11	0
	<sup>d</sup> Mean	78.2	88.6	79.8	
	Median	84.8	89.7	79.5	
	Minimum	42.6	55.0	67.6	
Laboratory reagent blanks for soils	N	17	13	15	14
	Mean	63.3	59.9	52.4	53.0
	Median	64.8	56.9	52.9	49.9
	Minimum	31.5	39.3	23.9	19.4
WIPP EM soils	N	120	91	112	112
	Mean	70.8	62.2	58.6	72.0
	Median	73.1	62.1	59.1	75.6
	Minimum	3.0	18.4	21.9	14.2
Replicate samples for soils	N	16	12	13	14
	Mean	71.9	61.5	52.8	68.8
	Median	72.3	61.6	47.2	80.2
	Minimum	47.3	38.7	21.7	12.2
Laboratory reagent blanks for drinking & surface water	N	3	3	3	3
	Mean	77.9	48.0	64.0	82.5
	Median	83.6	64.9	71.5	87.6
	Minimum	59.6	3.8	45.3	60.7
WIPP EM drinking & surface water	N	27	26	27	27
	Mean	80.1	68.1	69.7	90.5
	Median	82.5	81.3	74.7	94.4
	Minimum	37.0	2.3	28.1	40.3
Laboratory reagent blanks for FAS quarterly composite	N	2	2	2	2
	Mean	54.0	65.3	62.8	77.3
	Median	54.0	65.3	62.8	77.3
	Minimum	39.9	56.5	59.5	70.7
WIPP EM FAS quarterly composites	N	3	3	3	3
	Mean	83.2	80.3	69.0	85.0
	Median	86.1	78.0	76.8	80.1
	Minimum	71.7	74.9	52.9	75.9

<sup>a</sup>Tracer = An isotope of the radionuclide of interest, that is distinguishable from the analyte of interest, but assumed to behave the same in radiochemical processes

<sup>b</sup>EML-QAP = Environmental Measurement Laboratory Quality Assurance Program; WIPP EM = Waste Isolation Pilot Plant Environmental Monitoring project

<sup>c</sup>N = number of samples included in each analysis. EML and NRIP analyses did not require the determination of Th.

<sup>d</sup>Mean = Arithmetic mean

**Table L4. Mean Relative Percent Differences for Replicates in Radioanalyses of Actinides**

<sup>a</sup> Analytical Group	<sup>b</sup> N	<sup>c</sup> Relative Percent Difference								
		<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>241</sup> Am	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th
EML-QAP	75	3.4	5.9	2.5	1.9	6.9	1.9	<sup>d</sup> NA	NA	NA
WIPP EM Soils	106	<sup>e</sup> NC	16.2	20.8	9.7	14.2	7.8	5.6	7.5	5.6
<sup>f</sup> Percent Meeting ANSI 42.22 Standard for Precision										
EML-QAP	75	100	100	100	100	100	100	NA	NA	NA
WIPP EM Soils	106	<sup>e</sup> NC	100	100	69	100	69	79	79	86

<sup>a</sup>EML-QAP = Environmental Measurement Laboratory Quality Assurance Program; WIPP EM = Waste Isolation Pilot Plant Environmental Monitoring Pilot Plant

<sup>b</sup>N = number of analyses included in each analytical group

<sup>c</sup>Relative Percent Difference (RPD) = absolute value of difference between analyte concentrations in replicate/split samples, divided by the average of the two concentrations, expressed as a percent.

<sup>d</sup>NA = analyte not included in analytical group

<sup>e</sup>NC=cannot be calculated; one or both sample in all pairs had <sup>238</sup>Pu concentrations less than

<sup>f</sup>ANSI 42.22 Standard for Precision requires no significant difference between analytes at  $\alpha \approx 0.2$



## **GLOSSARY**

- aerosol - particles dispersed in a gas.
- aliquot - a subsample drawn from a larger sample.
- alpha-emitting – producing ionizing radiation in the form of alpha particles.
- anthropogenic - referring to environmental alterations resulting from the presence or activities of humans.
- actinides - the series of radioactive elements that starts with actinium and ends with lawrencium.
- aerodynamic diameter – distance around a sphere of unit density with the same settling velocity as the particle size of concern.
- attenuation - the reduction in level of a quantity, such as the intensity of a wave, over an interval of a variable, such as the distance from a source.
- coefficient of variation – a statistical parameter that expresses standard deviation as a percentage of the mean, calculated as standard deviation divided by the mean, multiplied by 100.
- Compton continuum - energy levels in the response of a photon spectroscopy instrument corresponding to scattered electrons.
- dosimetry - the measurement of radiation doses.
- gamma-emitting – producing ionizing radiation in the form of gamma rays.
- gross alpha - measurement of total number of alpha decays without specification of individual energies
- in vivo* - taking place in a living cell or organism.
- informatics - information management systems.
- kiloelectronvolts - a unit of energy, equal to 1,000 electronvolts.
- lumen – a unit of luminous flux equal to the light emitted in a unit solid angle by a uniform point source of one candle intensity.
- multi-channel analyzer – a device that convert successive electronic signals into parallel amplitude channels.
- photon - a massless particle, the quantum of the electromagnetic field, carrying energy, momentum and angular momentum.
- radionuclide - a type of atom that loses particles and energy through decay or transformation into other elements.
- standard deviation – a statistical parameter, calculated as the positive square root of the expected value of the square of the difference between a random variable and its mean.
- standard error – the standard deviation of the probability function or probability density function of a random variable and of a statistic; typically the standard deviation of the mean.
- temporal - pertaining to or limited by time.
- tertiary - third level.