

Long-Term Environmental Monitoring of an Operating Deep Geologic Nuclear Waste Repository - 8184

James Conca, Thomas Kirchner, James Monk, Sondra Sage
Carlsbad Environmental Monitoring and Research Center, IEE NMSU, 1400 University Drive, Carlsbad, NM 88220

ABSTRACT

In the present energy dilemma in which we find ourselves, the magnitude of humanity's energy needs requires that we embrace a multitude of various energy sources and applications. Nuclear energy must be a major portion of the distribution. One often-cited strategic hurdle to the commercial production of nuclear energy is the apparent lack of an acceptable nuclear waste repository. This issue has been quietly addressed at the U. S. Department of Energy (DOE) Waste Isolation Pilot Plant (WIPP; see <http://www.wipp.energy.gov>), the closest population center of significant size being Carlsbad, New Mexico. WIPP has been operating for about nine years, disposing of over 250,000 drum-equivalents of nuclear waste. From the standpoint of addressing operational and environmental risk, as well as public fear, WIPP has had extensive human health and environmental monitoring. The Carlsbad Environmental Monitoring and Research Center is in the Institute for Energy and the Environment, in the College of Engineering at New Mexico State University. Located in Carlsbad, NM, CEMRC has been the independent monitoring facility for the area around WIPP from 1993 to the present, i.e., from six years before disposal operations began to nine years of waste disposal operations (www.cemrc.org). Based on the radiological analyses of monitoring samples completed to date for area residents and site workers, and for selected aerosols, soils, sediments, drinking water and surface waters, there is no evidence of increases in radiological contaminants in the region of WIPP that could be attributed to releases from WIPP. Levels of radiological and non-radiological analytes measured since operations began in 1999 have been within the range of baseline levels measured previously, and are within the ranges measured by other entities at the State and local levels since well before disposal phase operations began in 1999.

INTRODUCTION

At present, WIPP is licensed to accept transuranic (TRU) waste generated from Defense activities. This waste must have activities exceeding 100 nanocuries per gram of waste from alpha-emitting transuranic isotopes with half-lives greater than 20 years, e.g., Pu, U, Am and Np. The dominant radionuclide in the waste from a performance standpoint is Pu. Nuclear waste first began being received at WIPP on March 26, 1999, consisting only of Contact Handled (CH), that is, drums with a surface exposures less than 200 mrem/hr, resulting primarily from alpha-emitting isotopes (Fig. 1). Mixed waste was first received by WIPP on September 9, 2000, and higher-activity waste (called remote handled or RH waste) was first received at the beginning of 2007. RH waste has surface exposures greater than 200 mrem/hr, so must be shielded and remotely handled (Fig. 2). It still must have activity concentrations greater than 100 nanocuries per gram of waste, with an upper limit of 23 Curie/liter. These higher activities result mostly from gamma emissions from the decay of isotopes such as ^{137}Cs and ^{90}Sr . This upper limit is similar to packaged processed high-level waste such as high level waste sludge or glass, and, in fact, was decided upon prior to the once-through fuel cycle decision of the 1970's. The RH waste is highly shielded, shipped in a 72B casket, and inserted remotely into a horizontal borehole in the disposal room wall. This borehole is a single drum-width in diameter and three drum-lengths deep with a shield plug, very similar geometrically to many high-level disposal strategies. DOE is currently investigating the use of shielded containers to emplace RH waste among the CH waste containers in the rooms of the repository.

WIPP ENVIRONMENTAL MONITORING

CEMRC was established in 1991 with a grant from the DOE with the primary mission being to develop and implement an independent health and environmental monitoring program in the vicinity of WIPP and to make the results easily accessible to all interested parties. The monitoring 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



Fig. 1. Over 10,000 CH nuclear waste drums and standard waste boxes emplaced within 1 of 56 rooms to be filled at WIPP over a 20-year period. 20 rooms have been filled as of this writing.



Fig. 2. Remote Handled nuclear waste (>100 nanoCi/gram of waste but <23 Ci/liter), some of it from reprocessing, being transported to the WIPP site in New Mexico in a 72B cask.

WIPP, although the quality assurance program at CEMRC allows the data to be used for many purposes. Analytical results and interpretations from the monitoring are published by CEMRC to inform the public and particularly the environmental science community.

Airborne particulate matter (aerosols) have been the major focus of the monitoring effort because, in the event that radioactive or chemical contaminants are released from WIPP, these materials could be rapidly dispersed through the atmosphere and spread throughout the environment. In addition, monitoring of the regional public through the Lie Down and Be Counted program (whole body counting) as well as workers is of the utmost importance as humans are the most important target whatever will be the transmission vector for contaminants. Sixteen soil sampling locations distributed around the repository provide geographic coverage for monitoring that is far greater than can be maintained for aerosol monitoring, albeit at a coarse temporal resolution. Drinking water, surface water and sediment monitoring are also conducted in the region around WIPP.

Past public surveys indicated that air monitoring and direct monitoring of people (whole body counting), followed by monitoring of drinking water, were the areas of greatest public interest. While it is highly unlikely that any chemical impacts of the WIPP will be detected through analyses of media other than air and people, CEMRC monitors soil, water, and sediment in some form and frequency decided with input from various stakeholders, in which one of the media other than air and people are sampled each year on a rotating basis. Until recently, vegetation and biota were also sampled. As an example, in 2005/2006 underground air exiting the repository, ambient air offsite, surface water and drinking water were sampled for various constituents and properties depending upon the media collected, and include gross alpha/beta, ^7Be , ^{212}Bi , ^{213}Bi , ^{214}Bi , ^{144}Ce , ^{249}Cf , ^{60}Co , ^{134}Cs , ^{137}Cs , ^{152}Eu , ^{154}Eu , ^{40}K , ^{233}Pa , $^{234\text{m}}\text{Pa}$, ^{212}Pb , ^{214}Pb , ^{106}Rh , ^{125}Sb , ^{208}Tl , ^{228}Ac , ^{234}U , ^{235}U , ^{238}U , ^{230}Th , ^{232}Th , ^{228}Th , ^{241}Am , ^{238}Pu and $^{239,240}\text{Pu}$. Inorganic constituents include those normally analyzed in waters (see Table 3).

The *in vivo* bioassay (whole body counting) program at CEMRC participates in the Department of Energy's *In Vivo* Laboratory Accreditation Program (DOELAP) via WIPP, and is currently accredited to perform the following direct bioassays:

- Transuranium elements via L x-ray in lungs,
- ^{241}Am , ^{234}Th , ^{235}U , fission and activation products in lungs including ^{54}Mn , ^{58}Co , ^{60}Co and ^{144}Ce , and
- Fission and activation products in total body including ^{134}Cs and ^{137}Cs (and ^{57}Co , ^{88}Y and ^{133}Ba).

The following sections show some long-term monitoring results for people, air, water and soil.

WHOLE BODY DOSIMETRY

Citizen volunteers from the Carlsbad area were monitored for internally deposited radionuclides through a project entitled *Lie Down and Be Counted* (LDBC). This project is an outreach service to the public and supports 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 arrival of the first waste shipment to WIPP serve as a baseline for comparisons with periodic follow-up measurements that are slated to continue throughout the operational phase of the repository. 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 detection limits and the status of the project and results are available in annual reports and on the CEMRC website (<http://www.cemrc.org>).

As of December 31, 2006, 783 individuals had participated in the LDBC project. At the time the WIPP opened, 366 individuals had been measured. This group of 366 measurements constitutes the pre-operational baseline to which subsequent results are compared. Counts performed after the opening of the WIPP are considered to be a part of the operational monitoring phase of the WIPP EM. Recounts began in July 1999, and 249 recount bioassays had been performed through December 2006. In addition, 217 new volunteers have participated in the program since October 1, 2002.

Table I: *Lie Down and Be Counted* Results through December 31, 2006

Radionuclide	<i>In Vivo</i> Count Type	Baseline Counts ^c (margin of error) (data prior to 27 March 1999) ^a N = 366	Operational Monitoring Counts (margin of error) (27 March 1999 – 31 December 2006) N = 690
		% of Results \geq ^b L _C	% of Results \geq L _C
²⁴¹ Am	Lung	5.2 (4.0 to 6.4)	4.1 (3.3 to 4.8)
¹⁴⁴ Ce	Lung	4.6 (3.5 to 5.7)	3.3 (2.6 to 4.0)
²⁵² Cf	Lung	4.1 (3.1 to 5.1)	5.8 (4.9 to 6.7)
²⁴⁴ Cm	Lung	5.7 (4.5 to 7.0)	4.6 (3.8 to 5.4)
¹⁵⁵ Eu	Lung	7.1 (5.8 to 8.4)	4.6 (3.8 to 5.4)
²³⁷ Np	Lung	3.6 (2.6 to 4.5)	4.2 (3.4 to 5.0)
²¹⁰ Pb	Lung	4.4 (3.3 to 5.4)	6.1 (5.2 to 7.0)
Plutonium Isotope	Lung	5.7 (4.5 to 7.0)	5.4 (4.5 to 6.2)
^d ²³² Th via ²¹² Pb	Lung	34.2 (31.7 to 36.6)	33.9 (32.1 to 35.7)
²³² Th	Lung	4.9 (3.8 to 6.0)	5.1 (4.2 to 5.9)
²³² Th via ²²⁸ Th	Lung	4.1 (3.1 to 5.1)	5.1 (4.2 to 5.9)
²³³ U	Lung	5.7 (4.5 to 7.0)	9.6 (8.4 to 10.7)
²³⁵ U/ ²²⁶ Ra	Lung	10.7 (9.0 to 12.3)	11.2 (10.0 to 12.4)
Natural Uranium via ²³⁴ Th	Lung	5.2 (4.0 to 6.4)	6.1 (5.2 to 7.0)
¹³³ Ba	Whole Body	3.6 (2.6 to 4.5)	3.0 (2.4 to 3.7)
¹⁴⁰ Ba	Whole Body	5.2 (4.0 to 6.4)	3.9 (3.2 to 4.7)
¹⁴¹ Ce	Whole Body	3.6 (2.6 to 4.5)	4.9 (4.1 to 5.8)
⁵⁸ Co	Whole Body	4.4 (3.3 to 5.4)	2.5 (1.9 to 3.1)
^d ⁶⁰ Co	Whole Body	54.6 (52.0 to 57.2)	28.6 (26.9 to 30.3)
⁵¹ Cr	Whole Body	5.7 (4.5 to 7.0)	4.2 (3.4 to 5.0)
¹³⁴ Cs	Whole Body	1.6 (1.0 to 2.3)	2.5 (1.9 to 3.1)
¹³⁷ Cs	Whole Body	28.4 (26.1 to 30.8)	21.2 (19.6 to 22.7)
¹⁵² Eu	Whole Body	7.4 (6.0 to 8.7)	6.2 (5.3 to 7.2)
¹⁵⁴ Eu	Whole Body	3.8 (2.8 to 4.8)	2.6 (2.0 to 3.2)
¹⁵⁵ Eu	Whole Body	3.8 (2.8 to 4.8)	3.8 (3.1 to 4.5)
⁵⁹ Fe	Whole Body	3.8 (2.8 to 4.8)	5.8 (4.9 to 6.7)
¹³¹ I	Whole Body	5.2 (4.0 to 6.4)	3.9 (3.2 to 4.7)
¹³³ I	Whole Body	3.3 (2.3 to 4.2)	4.1 (3.3 to 4.8)
¹⁹³ Ir	Whole Body	4.1 (3.1 to 5.1)	4.2 (3.5 to 5.0)
⁴⁰ K	Whole Body	100.0 (100.0 to 100.0)	100.0 (100.0 to 100.0)
^d ⁵⁴ Mn	Whole Body	12.3 (10.6 to 14.0)	11.9 (10.7 to 13.1)
¹⁰³ Ru	Whole Body	2.2 (1.4 to 3.0)	1.6 (1.1 to 2.1)
¹⁰⁶ Ru	Whole Body	4.4 (3.3 to 5.4)	3.9 (3.2 to 4.7)
¹²⁵ Sb	Whole Body	5.2 (4.0 to 6.4)	3.8 (3.0 to 4.5)
²³² Th via ²²⁸ Ac	Whole Body	34.7 (32.2 to 37.2)	25.2 (23.5 to 26.8)
⁸⁸ Y	Whole Body	7.7 (6.3 to 9.0)	6.0 (5.1 to 6.9)
⁹⁵ Zr	Whole Body	6.6 (5.3 to 7.9)	4.1 (3.3 to 4.8)

^a N = number of individuals. **Baseline counts include only the initial counts during this baseline period.**

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

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

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

Demographic characteristics of the LDBC cohort are generally consistent with those reported in the 2000 census for citizens living in Carlsbad. The largest deviation between the LDBC cohort and 2000 census is under-sampling of Hispanics. 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. Baseline monitoring includes only the initial count of individuals made prior to March 26, 1999. Operational monitoring includes the counting of new individuals and the recounting of previously measured participants. Based on all data collected, there is no evidence of an increase in the frequency of detection of internally deposited radionuclides for citizens living within the vicinity of the WIPP since the WIPP began receipt of radioactive waste. The criterion, L_C , was used to evaluate whether a result exceeds background, and the use of this criterion will result in a statistically inherent 5% false-positive error rate per pair-wise comparison (5% of all measurements will be determined to be positive when there is no activity present in the person).

Results are shown in Table I. For the baseline measurements (Table I, $N = 366$), the percentage of results greater than L_C were consistent with a 5% random false-positive error rate, at the 95% confidence level (1 to 9%), for all radionuclides except ^{232}Th via the decay of ^{212}Pb , $^{235}\text{U} / ^{226}\text{Ra}$, ^{60}Co , ^{137}Cs , ^{40}K , ^{54}Mn , ^{232}Th via the decay of ^{228}Ac . Five of these (^{232}Th via ^{212}Pb , ^{60}Co , ^{40}K , ^{54}Mn [^{228}Ac interference] and ^{232}Th [via ^{228}Ac]) are part of the shield-room background and positive detection is expected at low frequency. ^{40}K is a naturally occurring isotope of an essential biological element, so detection in all individuals is expected. ^{137}Cs and $^{235}\text{U} / ^{226}\text{Ra}$ 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.

For the operational monitoring counts (Table I, $N = 690$), the percentage of results greater than L_C were consistent with baseline at a 95% confidence level (margin of error), except for ^{60}Co and ^{232}Th (via ^{228}Ac). For these radionuclides, the percentage of results greater than L_C decreased relative to the baseline. This would be expected for ^{60}Co , since the radionuclide has a relatively short half life (5.2 years), and the content within the shield has decreased via decay by approximately 59% since the baseline phase of monitoring. The differences in ^{232}Th (via ^{228}Ac) results between the baseline and operational monitoring phase were also observed in 2001 and 2002 and are likely due to the replacement of aluminum (tends to contain Th and U) in some of the detector cryostat components with those manufactured from low radiation background steel.

Each subject must fill out an extensive questionnaire, therefore, various relationships can be determined with respect to whole body radiation, such as ethnicity, age, radiation work history, consumption of wild game, nuclear medical treatments and European travel. ^{40}K results were positive for all participants through December 2006 and ranged from 792 to 5558 Bq per person with an overall mean (\pm SE) of 2526 (\pm 26) Bq per person. Such results are expected since K is an essential biological element contained primarily in muscle, and a theoretical constant fraction of all naturally occurring K is the radioactive isotope ^{40}K . The mean ^{40}K value for males (\pm SE), was 3104 (\pm 30) Bq per person, which was significantly greater ($p < 0.0001$) than that of females, which was 1900 (\pm 22) Bq per person. This result was expected since, in general, males tend to have larger body sizes and greater muscle content than females.

Detectable ^{137}Cs is present in $23 \pm 3\%$ (95% confidence level, baseline and operational monitoring counts) of citizens living in the Carlsbad area. These results are consistent with worldwide general findings (Webb et al., 2000). Detectable ^{137}Cs body burdens ranged from 4.9 to 77.5 Bq per person with an overall mean (\pm SE) of 10.6 (\pm 0.6) Bq per person. The mean ^{137}Cs body burden for males (\pm SE), was 11.5 (\pm 0.8) Bq per person, which was significantly greater ($p = 0.002$) than that of females, which was 8.7 (\pm 0.3) Bq per person. As previously reported (CERMC Reports and [1]) the presence of ^{137}Cs was independent of ethnicity, age, radiation work history, consumption of wild game, nuclear medical treatments and European travel. However, the occurrence of detectable ^{137}Cs was associated with gender where males had higher prevalence of ^{137}Cs relative to females. Furthermore, the presence of ^{137}Cs was associated with smoking. Smokers had a higher prevalence of detectable ^{137}Cs (29.7 %) as compared to non-smokers (24.1 %). It is likely that the association with gender is related to the tendency for larger muscle mass in males than in females, as supported by the ^{40}K results. The association of ^{137}Cs with smoking could be related to the presence of fallout ^{137}Cs in tobacco, decreased pulmonary clearing capability in smokers, or other as yet unidentified factors.

These results, particularly the absence of detectable levels of plutonium, suggest that there have been no significant releases from WIPP. The percentage of results greater than L_C for $^{235}\text{U} / ^{226}\text{Ra}$ (11%) are significantly higher than the distribution-free confidence interval for a 5 % random false-positive error rate. These data are not nearly as compelling as those for ^{137}Cs , but the large sample size of the current cohort tends to support the observed pattern. Although ^{235}U and ^{226}Ra cannot be differentiated via gamma spectroscopy, it is likely the signal is the result of ^{226}Ra because the natural abundance of ^{226}Ra is much greater than that of ^{235}U .

AEROSOLS

CEMRC monitors two types of aerosols in the area of WIPP. Station A, an above-ground fixed air sampling platform, provides a way to monitor for releases of radionuclides and other substances in the exhaust air from the WIPP. Station A is located where radioactive or hazardous materials would most likely first be detected in the event of a release. CEMRC commenced sampling of the WIPP exhaust air at Station A on December 12 1998. The samples are collected on 47 mm diameter membrane filters with the use of a shrouded probe, commonly referred to as a fixed air sampler or FAS. The airflow through the FAS is approximately 170 liters per minute. The FAS sample filters are normally changed daily. All the analyses of the FAS filters are performed according to methods detailed in CEMRC document-controlled, standard operating procedures. After the samples are returned to the laboratory, the individual filters are first weighed to determine mass loadings, and after allowing for the decay of short-lived radon daughters, they are counted for gross alpha/beta activities for 1200 minutes using a low-background gas proportional counter (a Canberra LB4100 and, starting in 2006, a Protean MPC9604).

The gross alpha and beta activities shown here are expressed as *activity concentrations*, calculated as the activity per unit volume of air sampled (mBq m^{-3}). The minimum detectable activity concentrations for gross alpha were $\approx 0.1 \text{ mBq m}^{-3}$, while for gross beta is $\approx 0.2 \text{ mBq m}^{-3}$. Elemental and gamma-ray analyses are conducted on weekly composites of the filters. Individual FAS filters are digested using a mixture of strong acids in a microwave digestion unit, and weekly composites were prepared from the digestates of the individual filters. The weekly composites are then analyzed for a suite of trace elements with the use of a Perkin-Elmer Elan inductively coupled plasma-mass spectrometry (ICP-MS). The ICP-MS methods can provide data for up to ~ 35 elements, but in practice the concentrations of some elements, including As, Be, Cd, Er, Eu, Sc, Se, Sm, Tl and V are often below detectable or quantifiable levels, and a second set of elements (notably Ag, Li and Sn) has variable concentrations in blank filters which makes their quantification difficult. Analyses of gamma emitters are performed on the same weekly composites as used for the elemental studies; gamma analyses are done using a low-background, high-purity Ge well detector and a count time of 24 hours.

Finally, quarterly, or more recently, monthly composites are prepared from the weekly composites, and these are used for the determination of actinide activities. Only one half of the composite sample is normally used for the determination of the actinide activities. The remaining aliquot is archived. The composite sample is evaporated to dryness, and the residue is digested in perchloric acid to destroy the black residue, which consists mostly of diesel exhaust particulates. This process ensures that fluorine is completely removed and all traces of organic filter residue have been oxidized. The actinides are then separated as a group by co-precipitation on $\text{Fe}(\text{OH})_3$. After dissolution, Pu, U, and Am are separated by anion exchange and extraction chromatography, and the sample planchettes are finally prepared for alpha spectrometry using rare-earth micro-precipitation.

The essence of the strategic design for the monitoring program, including the studies at Station A, has been to compare pre- vs. post-disposal data. The first radioactive waste shipments were received at the WIPP on March 26, 1999, and this is considered the cut-off date separating the pre-disposal phase from the post-disposal or operational phase. The WIPP first received mixed waste on September 9, 2000, and therefore data for samples collected prior to that date compose a pre-mixed waste baseline for the elemental data while those collected afterwards are considered operational.

Fig. 3 shows the daily alpha and beta activity measurements for Station A from the end of 1998 to the end of 2006. Data points are distinguished by color, with red being pre-disposal, blue being operational, black being Station A backup results, and green being filters collected as part of a separate probe fouling study. The gross alpha and beta activities in the samples collected prior to the receipt of the first waste shipment represent the pre-disposal background, and the bulk of the activity in those samples was due to naturally occurring radioactive materials, specifically radon daughters. As shown in Fig. 3, the pre-operational gross alpha activity concentrations were high compared with the annual mean values for the next five years. Gross alpha and beta activities exhibit clear seasonal variability with peaks occurring in winter. An especially pronounced annual cycle in alpha activity concentrations, with high values in December and January and low values mid-year is seen in 2004 to 2005. After 2005, alpha activities appear to have gone back up to pre-operational levels, while beta remains slightly lower than pre-operational levels.

Fig. 3 also shows the sensitivity of the monitoring program at Station A that was demonstrated in January 2001 when CEMRC found two samples with elevated gross beta activity concentrations in the Station A sample filters. Follow-up investigations eventually traced the source of the beta emitters to the discharge of a fire extinguisher underground, but the incident was more notable because it demonstrated for the first time the ability of the monitoring system to detect a non-routine event. A second, more significant incident occurred when scientists from CEMRC reported that they had detected a small quantity of Pu in a composite aerosol sample from the second calendar quarter of 2003. This discovery was later corroborated by other site monitoring programs through the analyses of samples that were independently collected and

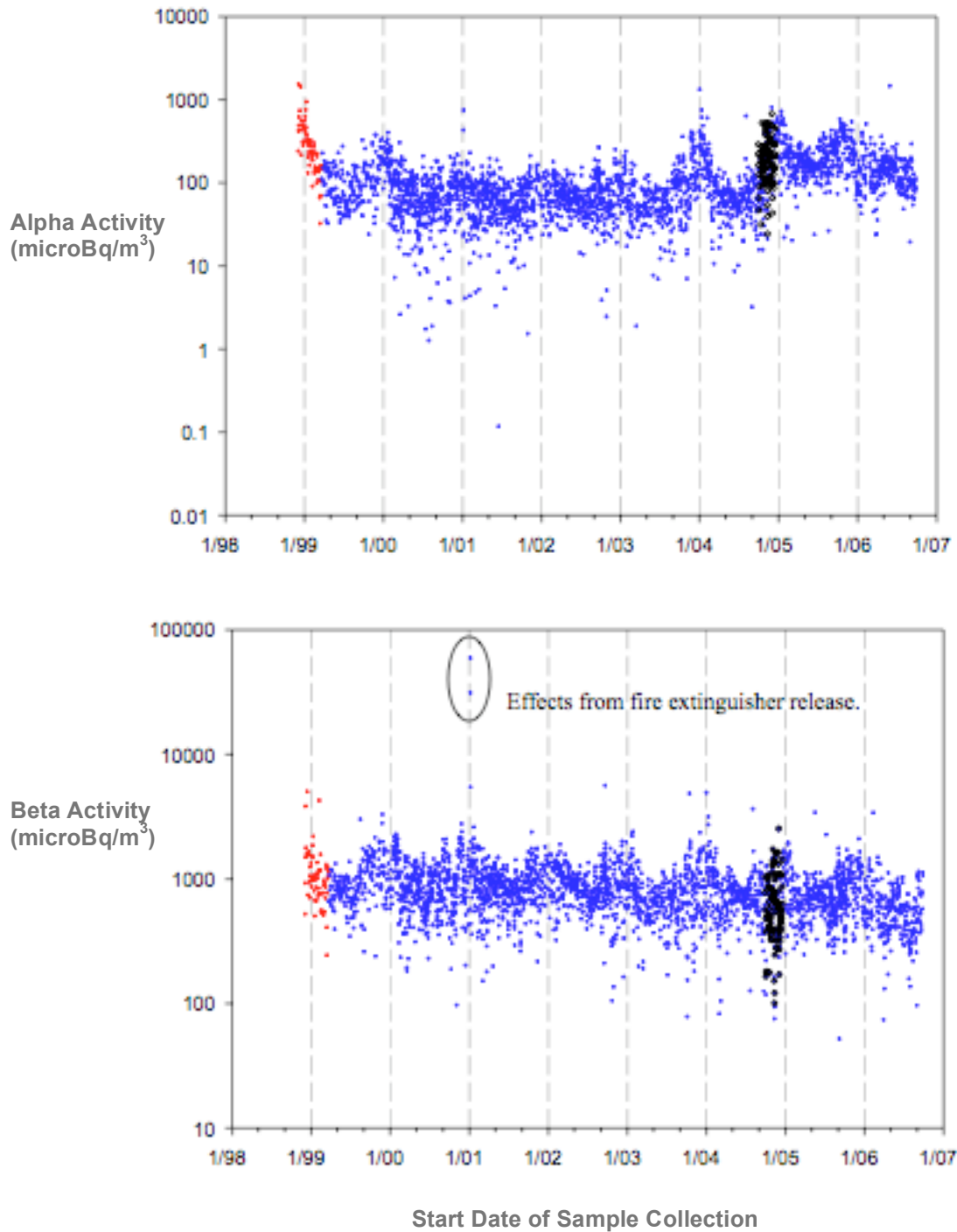


Fig. 3. Timeseries plots of gross alpha and gross beta activity concentrations of aerosols on filters sampling air exiting the WIPP underground (Station A FAS). Red points denote pre-disposal samples, blue points are for operational samples and black points are for samples collected with the back-up sampler.

analyzed. The activity was extremely low and well-within historic ambient air background, but indicated the ability of the monitoring program to detect radionuclides of interest at any level above the MDC.

Aerosol particle sampling is also conducted at three other sites: 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), collected using high-volume samplers (flow rate $\sim 1.13 \text{ m}^3 \text{ min}^{-1}$) for collecting total suspended particulate matter. Of particular interest to WIPP is Pu, which has been dispersed throughout the global environment mainly by nuclear weapons tests. When quantified by alpha spectrometry ^{239}Pu typically is determined together with ^{240}Pu , because isotopes are difficult to separate chemically, and they are represented as $^{239,240}\text{Pu}$ (^{239}Pu half-life, $t_{1/2} = 24,110 \text{ yr}$ and ^{240}Pu $t_{1/2} = 6563 \text{ yr}$). ^{239}Pu and ^{240}Pu also have similar alpha particle energies, about 5.25 MeV. Fig. 4 shows the results for $^{239,240}\text{Pu}$ from these aerosol collection stations. There is no apparent correlation between Pu and proximity to WIPP or beginning of WIPP operations. In fact, the more distant, upwind Cactus Flats site has generally higher activities, although all values are extremely low and in keeping with fallout as the source with a strong seasonal trend. Similar results were obtained for ^{241}Am . An important finding has been that the activity of Pu (and Am) and the concentration of Al in aerosols are correlated, driven by the resuspension of dust particles contaminated with radioactive fallout from past nuclear weapons tests [2].

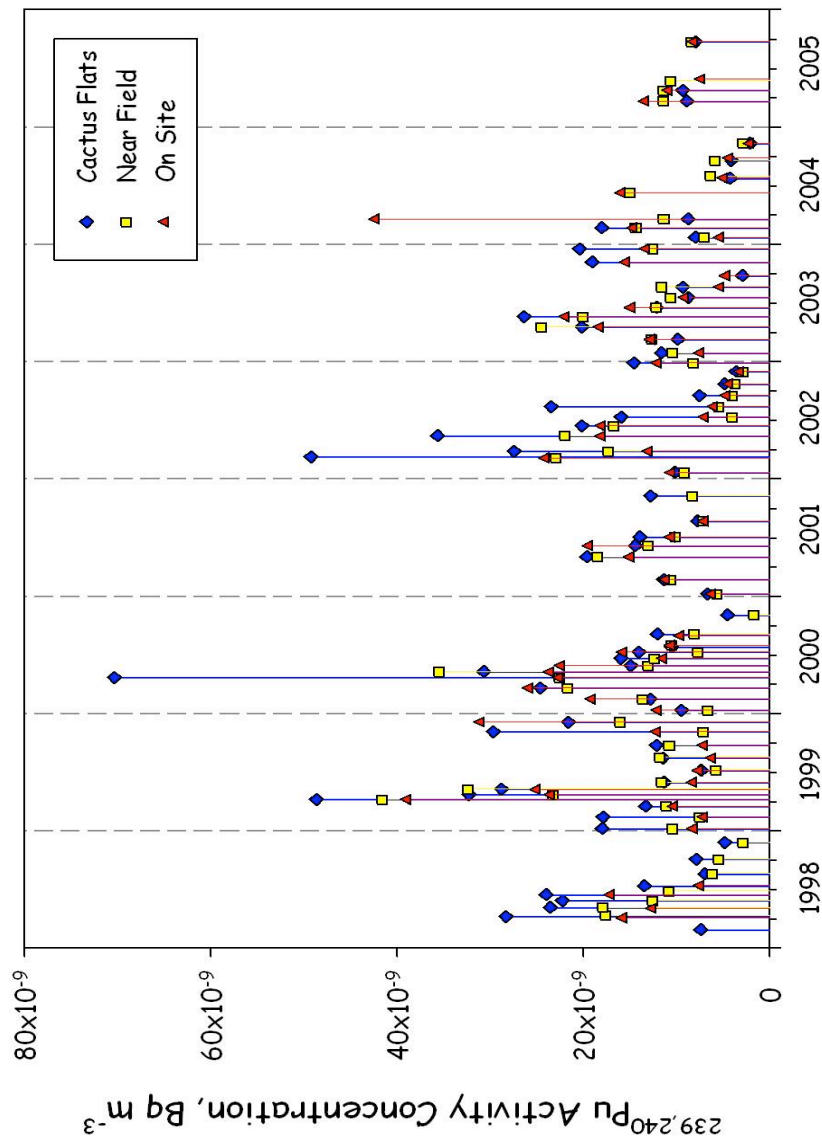


Fig. 4. High Volume Ambient Aerosol $^{239,240}\text{Pu}$ Activity Concentration at three sampling stations in and around WIPP.

REGIONAL DRINKING WATER

The monitoring program also focuses on the major drinking water supplies used by communities in the WIPP region because these are often perceived by the public as a potential route for contaminants to reach humans. Five community supplies of drinking water (representing three major regional aquifers) are included in routine sampling, including Carlsbad, Loving/Malaga, Otis, Hobbs and a secondary source for Carlsbad. One private water well (representing a fourth aquifer) that is located within 16 km of the WIPP is also sampled. Results have been generally consistent for each source across sampling periods, with few organic contaminants detected and inorganic substances mostly below levels specified under the Safe Drinking Water Act.

Drinking water samples were collected (and preserved) as follows: (1) 4 L for radiological analyses, (2) 1 L for elemental analyses and (3) 1 L for anion tests. None of the samples were filtered before analysis, but a portion of the 4 L sample was transferred to a 3 L Marinelli beaker for gamma spectroscopy analyses. Non-radiological analyses of drinking water samples were performed using ICP-MS. Samples were first counted in Marinelli beakers using a coaxial, high purity Ge detector system to determine gamma-emitting radionuclide activity concentrations. Radiochemistry was then applied to each sample for actinide separation and purification using multiple precipitation, co-precipitation and ion-exchange and/or extraction chromatography. Once the actinides were separated elementally, they were co-precipitated with LaF_3 and deposited onto filters, which were then counted on an alpha spectroscopy system. Aliquots were blank-corrected after application of dilution factors. In cases where blank corrections lowered solution concentrations below MDC values, concentrations greater than zero are reported; negative concentrations are reported as $< \text{MDC}$.

No radionuclides were measured above MDC in any drinking water samples as measured by gamma spectroscopy targeting 11 naturally occurring and 12 anthropogenic gamma-emitters. Three naturally occurring uranium isotopes (^{234}U , ^{235}U and ^{238}U) were detected in all drinking water samples as is the case for most waters in New Mexico (Table II). Levels are similar for the last eight years and within about a factor of two of the 1998 baseline levels. These levels and ratios of uranium are typical of natural variations in ground water [3,4] and agree well with the few directly comparable values reported from studies in the region. The federal and state action level for gross alpha emitters, which includes isotopes of Pu and U, is 15 pCi/L (0.56 Bq/L). This is over 10,000 times the levels measured by CEMRC in any drinking water sample over the last eight years. Neither $^{239,240}\text{Pu}$ or ^{238}Pu have been measured above the MDC in any drinking water samples, an example of which is given in Fig. 5 for $^{239,240}\text{Pu}$ in Carlsbad drinking water from 1998 to 2005.

Measurements of inorganic analytes by CEMRC from the five drinking water sources showed little variation between years for each source (Table III). Differences of a factor of two or three between one set of successive years is common. One important constituent in all drinking waters from the southwest is arsenic (As). As levels were in the expected range for all samples and all sites, but were as high as $7.8 \mu\text{g L}^{-1}$, suggesting that some of these these drinking water sources may exceed any As standard $\leq 5.0 \mu\text{g L}^{-1}$ as has been recently adopted by EPA but not yet enforced for many small municipalities. Arsenic in Carlsbad drinking water has consistently been between $0.3 - 0.7 \mu\text{g L}^{-1}$. As per the grant requirements and the fact that CEMRC does not use EPA compliance procedures, these results are not appropriate for use in assessing regulatory compliance. However, CEMRC results for drinking water collected for the last eight years agreed well with, and were generally below, measurements for the same elements published by the City of Carlsbad Municipal Water System (2005 *Annual Consumer Report on the Quality of Your Drinking Water* (www.cityofcarlsbadnm.com/documents/CCR2005.pdf)).

SUMMARY

The Carlsbad Environmental Monitoring and Research Center has been monitoring the region around WIPP, the deep geologic nuclear repository, from before disposal operations began to the present, about eight years of waste disposal operations. Based on radiological analyses of area residents and site workers, aerosols, soils, sediments, drinking water and surface waters, there is no evidence of increased radiological contaminants in the region of WIPP that could be attributed to releases from WIPP. Levels of radiological and non-radiological analytes measured since operations began in 1999 have been within the range of baseline levels measured previously, and are within the ranges measured by other entities at the State and local levels since well before disposal phase operations began in 1999.

Acknowledgments

The authors wish to thank Kim Ui Chearnaigh and Lisa Hudston for scientific and technical assistance, and Angela Najera for editing. This paper, like so much at CEMRC, is in memory of Barry Stewart who is dearly missed.

Table II. Range of Activity Concentrations for Uranium Isotopes Measured in Drinking Water During 2005 and Comparisons with 1998/1999 Baseline

Location	Analyte	2005 Activity Concentration (Bq L ⁻¹)		^a RPC from baseline (%)	
		Activity	Uncertainty	2005:1998	2005:1999
Carlsbad	²³⁴ U	2.75E-02	8.16E-04	-17.7	-6.5
	²³⁵ U	1.51E-03	1.41E-04	100.8	116.0
	²³⁸ U	1.11E-02	3.91E-04	-17.8	-2.6
Double Eagle	²³⁴ U	5.83E-02	1.60E-03	5.2	-5.8
	²³⁵ U	1.54E-03	1.43E-04	42.6	40.0
	²³⁸ U	2.48E-02	7.46E-04	22.2	6.9
Hobbs	²³⁴ U	9.82E-02	2.61E-03	11.1	15.0
	²³⁵ U	2.68E-03	2.11E-04	20.2	8.9
	²³⁸ U	4.27E-02	1.20E-03	11.5	10.6
Loving	²³⁴ U	8.11E-02	2.17E-03	-0.5	-4.3
	²³⁵ U	1.74E-03	1.55E-04	4.8	15.2
	²³⁸ U	2.63E-02	7.82E-04	8.7	0.0
Otis	²³⁴ U	1.17E-01	3.09E-03	-9.3	-22.0
	²³⁵ U	2.60E-03	2.07E-04	-4.8	-8.8
	²³⁸ U	4.36E-02	1.23E-03	-6.6	-17.7

^a RPC = relative percent change; for 2005:1998 = ((2005 concentration – 1998 concentration) / 1998 concentration) * 100%; for 2005:1999 = ((2005 concentration – 1999 concentration) / 1999 concentration) * 100%

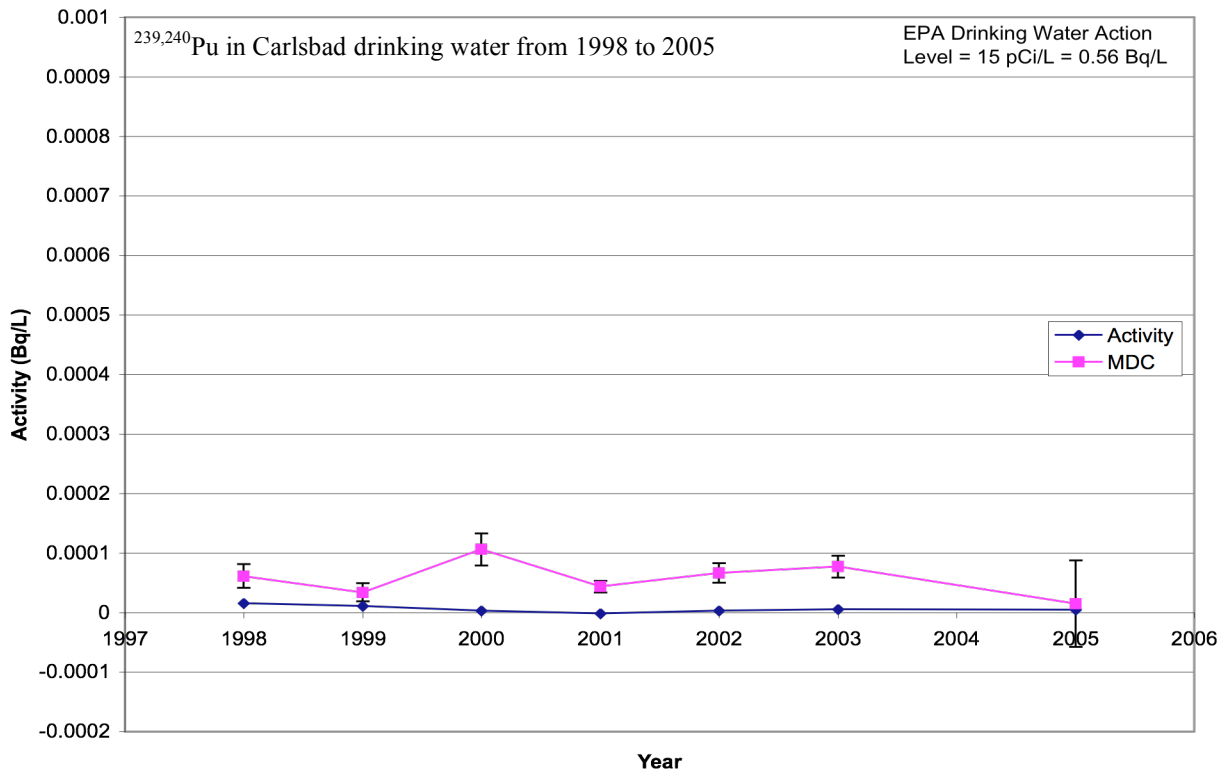


Fig. 5. ^{239,240}Pu in Carlsbad drinking water from 1998 to 2005. MDC is the minimum detection concentration, and all samples are below this lower limit. EPA Action levels are for all alpha-emitters, including U+Pu.

Table III. Selected Inorganic Analytes in Drinking Water from 1998 to 2005 at Five Locations

Carlsbad													
1998-2005					2003				2005				
EL ¹	N ²	N _{DET} ²	Min ³	Max ³	MDC ⁴ (ug/L)	Blank Conc. (ug/L)	Avg Conc. w/Blank Subt ⁵ (ug/L)	Avg Conc. w/o Blank Subt ⁵ (ug/L)	MDC ⁴ (ug/L)	Blank Conc. (ug/L)	Avg Conc. w/Blank Subt ⁵ (ug/L)	Avg Conc. w/o Blank Subt ⁵ (ug/L)	
Ag	8	2	1.23E-02	1.75E-02	3.00E-02	-1.36E-03	<MDC	<MDC	2.30E-02	1.11E-01	<MDC	<MDC	
Al	9	4	2.34E+00	3.17E+01	2.20E+00	-9.12E-01	<MDC	<MDC	1.49E+01	-2.80E+01	<MDC	<MDC	
As	9	6	3.45E-01	6.82E-01	1.80E+00	-3.04E-01	<MDC	<MDC	2.01E+00	3.19E+01	<MDC	<MDC	
B	1	1	3.07E+01	3.07E+01	3.40E+00	-1.23E+00	3.07E+01	3.07E+01	N/A ⁶	N/A	N/A	N/A	
Ba	9	9	6.64E+01	7.62E+01	1.40E-01	-1.63E-02	6.77E+01	6.77E+01	1.24E-01	-5.63E+00	7.62E+01	7.62E+01	
Be	8	0	N/A	N/A	8.00E-01	1.02E-02	<MDC	N/A	N/A	N/A	N/A	N/A	
Ca	8	8	6.54E+04	8.06E+04	2.40E+03	-1.47E+00	6.54E+04	6.54E+04	N/A	N/A	N/A	N/A	
Cd	8	0	N/A	N/A	3.20E-02	-1.51E-02	<MDC	<MDC	6.73E-02	1.91E+00	<MDC	<MDC	
Ce	7	0	N/A	N/A	1.20E-02	1.35E-03	<MDC	<MDC	N/A	N/A	N/A	N/A	
Co	8	6	8.80E-02	3.41E-01	5.00E-02	-3.07E-04	<MDC	<MDC	7.21E-01	8.25E-01	<MDC	<MDC	
Cr	9	7	2.68E+00	7.15E+00	5.60E-02	-8.89E-02	3.31E+00	3.22E+00	7.82E+00	9.24E+01	<MDC	<MDC	
Cu	9	8	1.23E+00	1.67E+01	3.10E-01	4.75E-02	2.11E+00	2.11E+00	8.98E-02	3.68E+01	1.67E+01	1.67E+01	
Dy	8	0	N/A	N/A	3.50E-02	3.30E-03	<MDC	<MDC	2.79E-02	-1.77E-01	<MDC	<MDC	
Er	8	0	N/A	N/A	2.70E-02	-1.40E-03	<MDC	<MDC	1.21E-02	-5.61E-02	<MDC	<MDC	
Eu	8	6	1.35E-02	2.43E-02	2.30E-02	-9.25E-04	<MDC	<MDC	1.64E-02	-1.04E-01	<MDC	<MDC	
Fe	9	3	2.14E+01	3.85E+01	7.60E+00	-1.51E+01	3.85E+01	2.34E+01	2.94E+02	3.66E+03	<MDC	<MDC	
Ga	2	2	3.24E+00	3.25E+00	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	
Gd	6	0	N/A	N/A	3.40E-02	-5.68E-04	<MDC	<MDC	N/A	N/A	N/A	N/A	
Hg	7	0	N/A	N/A	1.21E-02	9.60E-04	<MDC	<MDC	N/A	N/A	N/A	N/A	
K	8	8	1.04E+03	3.56E+03	4.40E+01	-1.16E+01	1.29E+03	1.29E+03	N/A	N/A	N/A	N/A	
La	8	5	1.41E-02	4.48E-02	1.50E-02	-5.90E-04	<MDC	<MDC	1.36E-02	1.00E-01	<MDC	<MDC	
Li	7	7	6.09E+00	7.87E+00	4.00E-01	-1.12E-02	6.14E+00	6.14E+00	N/A	N/A	N/A	N/A	
Mg	8	8	3.14E+04	3.47E+04	1.20E+01	2.01E-01	3.20E+04	3.20E+04	5.97E+00	-8.94E+01	3.47E+04	3.47E+04	
Mn	9	8	5.50E-02	9.40E-01	1.66E-01	1.65E-02	5.31E-01	5.31E-01	1.60E+00	1.97E+01	<MDC	<MDC	
Mo	9	8	7.03E-01	1.26E+00	1.10E-01	-9.78E-04	1.20E+00	1.20E+00	1.35E+00	-3.05E+01	<MDC	<MDC	
Na	8	8	1.28E+04	9.94E+04	1.12E+03	-3.13E+00	2.09E+04	2.09E+04	N/A	N/A	N/A	N/A	
Nd	8	0	N/A	N/A	3.40E-02	5.32E-03	<MDC	<MDC	1.65E-02	3.60E-01	<MDC	<MDC	
Ni	8	7	1.01E+00	2.89E+00	1.90E-01	-9.44E-03	1.01E+00	1.01E+00	1.74E+00	1.14E+01	<MDC	<MDC	
P	1	0	N/A	N/A	N/A	N/A	N/A	N/A	2.27E+01	-2.59E+02	<MDC	<MDC	
Pb	8	8	1.63E-01	8.53E+00	3.50E-02	1.11E-02	6.02E-01	6.02E-01	3.16E-02	-7.17E+00	8.53E+00	1.36E+00	
Pr	8	0	N/A	N/A	1.20E-02	6.91E-04	<MDC	<MDC	1.29E-02	-1.81E-01	<MDC	<MDC	
Rh	2	1	1.10E-02	1.10E-02	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	
Sb	9	4	3.00E-02	1.99E-01	2.50E-02	-1.86E-04	3.00E-02	3.00E-02	1.02E-01	1.96E-01	<MDC	<MDC	
Sc	6	6	1.72E+00	3.11E+00	8.20E-01	-2.93E-01	1.90E+00	1.90E+00	N/A	N/A	N/A	N/A	
Se	8	3	9.25E-02	1.75E+00	1.90E+00	6.50E-01	<MDC	<MDC	N/A	N/A	N/A	N/A	
Si	4	4	5.31E+03	1.68E+04	1.00E+03	-1.29E-01	5.31E+03	5.31E+03	7.32E+01	-1.01E+04	1.68E+04	6.77E+03	
Sm	8	7	2.34E-02	3.64E-02	3.00E-02	-3.80E-03	3.24E-02	3.24E-02	1.62E-02	-2.01E-01	<MDC	<MDC	
Sn	5	1	5.97E-02	5.97E-02	5.50E-02	-3.96E-03	<MDC	<MDC	N/A	N/A	N/A	N/A	
Sr	9	9	3.23E+02	4.59E+02	1.80E-01	2.11E-02	3.54E+02	3.54E+02	2.51E-01	-1.19E-01	3.48E+02	3.48E+02	
Th	8	1	1.98E-02	1.98E-02	5.40E-03	-6.30E-05	<MDC	<MDC	1.49E-02	6.30E-03	<MDC	<MDC	
Ti	4	3	3.64E-01	4.22E+00	2.60E+00	-1.00E-01	4.22E+00	4.22E+00	N/A	N/A	N/A	N/A	
Tl	8	8	9.97E-02	1.54E-01	1.20E-02	-2.21E-04	1.47E-01	1.47E-01	1.07E-02	2.05E-02	9.97E-02	9.97E-02	
U	8	8	8.21E-01	8.79E-01	8.60E-03	-5.83E-04	8.79E-01	8.79E-01	7.76E-03	-9.50E-04	8.60E-01	8.60E-01	
V	9	9	3.82E+00	5.90E+00	2.80E+00	3.16E-01	4.91E+00	4.91E+00	2.75E+00	-2.71E+00	4.53E+00	4.53E+00	

¹El = Element analyzed; ²N = Total number of samples analyzed; N_{det} = number of samples with detectable (above MDC) values; ³Min = the lowest value measured above MDC; Max = the highest value measured; ⁴MDC = Minimum detectable concentration; ⁵Average sample values with and without subtraction of the blank value; when blank subtraction is performed, it is only done when the blank value falls outside of the range (-MDC < blank < +MDC) ⁶N/A = Not Applicable

REFERENCES

1. J.L. WEBB and T.B. KIRCHNER. "An evaluation of in vivo sensitivity via public monitoring." Radiation Protection Dosimetry, 89 (3-4): 183-191 (2000).
2. T.B. KIRCHNER, J.L. WEBB, S.B. WEBB, R. ARIMOTO, D.A. SCHOEP, and B.D. STEWART. "Variability in background levels of surface soil radionuclides in the vicinity of the WIPP." J. of Env. Radioactivity 60:275-291 (2002).
3. C.R. COTHERN and W.L. LAPPENBUSCH. "Occurrence of uranium in drinking water in the US." Health Physics, 45:89-99 (1983).
4. S. LUO, T. KU, R. ROBACK, M. MURRELL, and T. MCLING. "In-situ radionuclide transport and preferential groundwater flows at INEEL (Idaho)." Geochim. Cosmochim., 64 No. 5:867-881 (2000).