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PAPER

Plutonium in the WIPP environment: its detection, distribution and behavior

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The Waste Isolation Pilot Plant (WIPP) is the only operating deep underground geologic nuclear repository in the United States. It is located in southeastern New Mexico, approximately 655 m (2150 ft) below the surface of the Earth in a bedded Permian evaporite salt formation. This mined geologic repository is designed for the safe disposal of transuranic (TRU) wastes generated from the US defense program. Aerosol and soil samples have been collected near the WIPP site to investigate the sources of plutonium in the WIPP environment since the late 1990s, well before WIPP received its first shipment. Activities of ^{238}Pu, $^{239+240}$Pu and ^{241}Am were determined by alpha spectrometry following a series of chemical separations. The concentrations of Al and U were determined in a separate set of samples by inductively coupled plasma mass spectrometry. The annual airborne concentrations of $^{239+240}$Pu during the period from 1998 to 2010 show no systematic interannual variations. However, monthly $^{239+240}$Pu particulate concentrations show a typical seasonal variation with a maximum in spring, the time when strong and gusty winds frequently give rise to blowing dust. Resuspension of soil particles containing weapons fallout is considered to be the predominant source of plutonium in the WIPP area. Further, this work characterizes the source, temporal variation and its distribution with depth in a soil profile to evaluate the importance of transport mechanisms affecting the fate of these radionuclides in the WIPP environment. The mean ^{137}Cs/$^{239+240}$Pu, ^{241}Am/$^{239+240}$Pu activity ratio and ^{240}Pu/^{239}Pu atom ratio observed in the WIPP samples are consistent with the source being largely global fallout. There is no evidence of any release from the WIPP contributing to radionuclide concentrations in the environment.

Introduction

The Waste Isolation Pilot Plant permanently disposes of transuranic (TRU) wastes that were generated from research and production of nuclear weapons at various Department of Energy sites. The facility is located in the remote Chihuahuan desert of southeastern New Mexico near Carlsbad. This site was chosen for its geological and physiological characteristics. The area sits on top of massive beds of salt. The disposal rooms are mined 665 metres (2150 feet) underground in a 700 metre thick salt formation that has been stable for more than 200 million years. As shown in Fig. 1, the WIPP repository consists of eight panels, each consisting of seven waste disposal rooms approximately 300 feet (91 metres) long and 33 feet (10 metres) wide. Seven disposal panels have been excavated, and the first five have been closed and sealed from ventilation air. Disposal is continuing in the sixth panel. The facility also consists of above ground waste receipt and handling facilities, common access drifts for access and ventilation to the disposal panels, and the four shafts connecting surface operations to underground emplacement activities. Disposal in salt was chosen mainly because massive salt deposits have extremely low hydraulic conductivity, *i.e.* no

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Environmental impact

The present work describes the detection, distribution and behavior of plutonium in the WIPP environment. The Waste Isolation Pilot Plant (WIPP) is the only operating deep underground geologic nuclear repository in the United States. This mined geologic repository is designed for the safe disposal of transuranic (TRU) wastes generated from the US defense program. The source and detection of radionuclides in and around the WIPP site are important to maintain the integrity of the site and to protect the public and environment. After twelve years of continuous operations, there is no evidence of increases in radiological contaminants in the region that could be attributed to releases from WIPP.

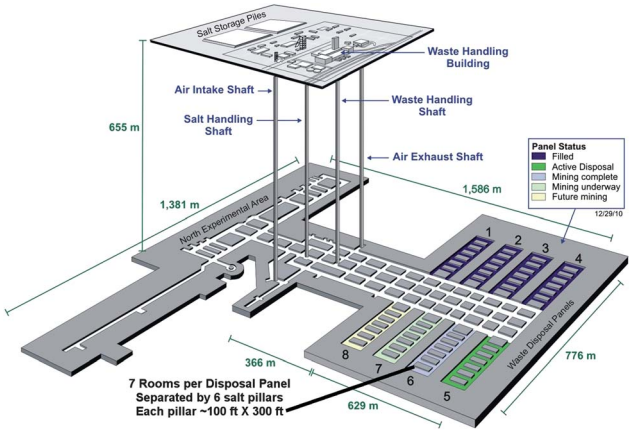

Fig. 1 WIPP layout.

flowing water that could move the waste out of the repository environs and to the surface. Salts are also relatively easy to mine. Additionally, rock salt heals its own fractures because of its plasticity and will slowly and progressively move in to fill the mined area and safely seal the radioactive waste from the environment. TRU waste is defined as radioactive waste containing more than 100 nanocuries (3700 becquerels [Bq]) of alpha-emitting TRU isotopes per gram of waste, with half-lives greater than twenty years. Most TRU waste is contaminated industrial trash, such as rags and old tools, sludges from solidified liquids, glass, metal, and other materials. TRU waste disposal operations began in 1999 and since then over 80 000 cubic metres of TRU waste in over 157 000 containers have been emplaced in the repository.

Since the air in the repository exits to the surface through the exhaust shaft, this shaft is the sole potential pathway for airborne radioactivity release from the WIPP during the operational phase. The potential for release is mitigated by HEPA (High Efficiency Particulate Air) filters at the surface. Continuous air monitors underground are used to control whether the ventilation returning to the surface is passed through these large filter systems before it is released to the atmosphere. In addition, there is some potential for radioactivity release if a drum was punctured or dropped during the emplacement operations. A less likely scenario is fire on one of the diesel-fueled vehicles used to transport waste containers from unloading areas to emplacement locations. A fuel-pool fire is the worst-case accident scenario analyzed in the WIPP Safety Analysis Report. The human intrusion scenario, after the loss of active institutional control (assumed to occur more than 300 years in the future), is the only credible release pathway from WIPP after the repository is closed. While a very small number of minor container handling incidents have occurred, none are known to have resulted in any external release of radioactive materials. It is important to note that these accident scenarios are very unlikely. The WIPP Technical Safety Requirements (TSR) impose limits on operations and waste handling to prevent release scenarios.

The vast majority of radionuclides within TRU waste are ^{239}Pu, ^{240}Pu and ^{241}Am, which account for more than 99% of the total radioactivity for most of the 10 000-year regulatory period. In this context, the variation in concentrations of these radionuclides in the WIPP environment is important not only because they are the main component of the WIPP wastes, but also because of their global background activity.

Atmospheric nuclear tests have been the major source of radiological contamination to date in the global environment. Approximately 6 tons of ^{239}Pu were introduced into the environment from more than 500 atmospheric weapon tests conducted between 1945 and 1980.[1] The fallout was distributed globally at an approximately 3 : 1 ratio between the northern hemisphere and the southern hemisphere. Additionally, local and regional contaminations of plutonium in the environment have resulted from nuclear accidents such as Chernobyl and Fukushima and around former nuclear weapons production sites both in the US and the former Soviet Union.

Currently, ^{238}Pu, ^{239}Pu and ^{240}Pu isotopes can be measured as traces in environmental samples with a ^{238}Pu/$^{239+240}$Pu activity ratio of 0.03 at mean latitudes of 40°–50° N tracing their global origin (UNSCEAR, 2000).[2] At present, almost all plutonium being introduced into the atmosphere can be found in the surface soil. Plutonium can migrate vertically at various rates depending on meteorological conditions, physiochemical properties of soil, and human activity. It can also be taken up by plants or resuspended into the air with eroded soil particles. The importance of resuspension in recycling radionuclides from soil back into the atmosphere has been pointed out in many publications.[3–6] In an effort to better understand the transport of these radionuclides and to document the levels of fallout of contaminants, a number of national and international monitoring programs were established. The fallout plutonium deposition up to the end of 1980 has been reported by Miyake *et al.*[7,8] and Katsuragi *et al.*[9] Hirose *et al.*[10] presented plutonium deposition at the MRI in Tsukuba, Japan to trace the fallout plutonium concentrations from 1957 to the end of 2001.

In the Carlsbad area, where WIPP is located, there is an additional potential local source of anthropogenic radioactivity from an underground nuclear test during the Plowshare project. One particular test occurred at the Gnome site, about 8.8 km southwest of the WIPP site, in 1961 when an underground test of a 3.3 kiloton ^{239}Pu device vented radioactive materials to the surface.[11] Clean-up efforts at this site have been carried out in several campaigns since that time, and the surface contamination is now well below the risk-based action levels. However, ^{137}Cs and plutonium have been detected in some samples of surface soils at the Gnome site.[12] These contaminated soils are of practical concern because they are a potential source of contamination for environmental samples being collected to monitor potential release of radionuclides from the WIPP. Consequently, it is very important to understand factors controlling the distribution of contaminants in the WIPP area.

In this article, the current trend of the radionuclides, specifically Pu and Am, is evaluated in the vicinity of the WIPP. The data reported were generated as part of the DOE sponsored WIPP environmental monitoring program by an independent oversight organization, the Carlsbad Environmental Monitoring & Research Center (CEMRC). The CEMRC was established in 1991 to conduct independent monitoring of the WIPP site. Results from this program are accessible to the public and are used here in evaluating the long-term history of these radionuclides to assess the impact of WIPP (if any) on the local environment because the public needs to know what is truly

happening in the environment and what effect WIPP operations have on their lives and health. Also, this type of information is important for assessing the impact of the WIPP on the local environment and for public acceptance of such projects.

Experimental

Gamma analysis was conducted using a high purity (HPGe) detector. A set of soil matrix standards procured from Eckert and Ziegler, Analytics (GA) was used to establish matrix-specific calibration and counting efficiencies. The minimum detectable activity for ^{137}Cs was 0.12 Bq kg^{-1} for a counting time of 48 hours. Alpha emitting isotopes of Pu and Am were measured for at least 5 days on alpha spectrometers (Oxford Oasis System, Oxford Instruments Inc.) with 72 vacuum chambers (PIPS detector with 22% efficiency, 20 keV Full Width-Half Maximum (FWHM), and 450 mm^2 active areas) after radiochemical separation/purification and micro-coprecipitation. The ^{240}Pu/^{239}Pu atom ratios were determined using Accelerator Mass Spectrometry (AMS) (McAninch 2000) at Lawrence Livermore National Laboratory (LLNL).

Air sample location and pre-treatment

Ambient aerosols are collected using high volume samplers ("hivols", flow rate ~1.13 m^3 min^{-1}) from three monitoring stations: (1) OnSite, which is about 0.1 km northwest (down prevailing wind) of the WIPP exhaust shaft, (2) Near Field, about 1 km northwest of the facility; and (3) Cactus Flats, about 19 km southeast (up prevailing wind) of the WIPP site. Aerosol sampling locations on and near the WIPP site are shown in Fig. 2. These sites were selected based on an analysis of the most probable scenario for radioactivity release if there was an accidental release during the operation of the WIPP. Each filter is weighed before and after sampling to determine the weight of solid material collected on the filters. The aerosol samples were collected on 20 × 25 cm A/E™ glass fiber filters (Pall German Laboratory, Ann Arbor, MI) taken over a period of 3 to 6 weeks depending on the levels of particulate matter that accumulated on the filters. For radiochemical analyses, filters are ashed in a muffle furnace at 500 °C for 6 hours and then each filter is digested with strong acid mixtures, HCl + HF + $HClO_4$.

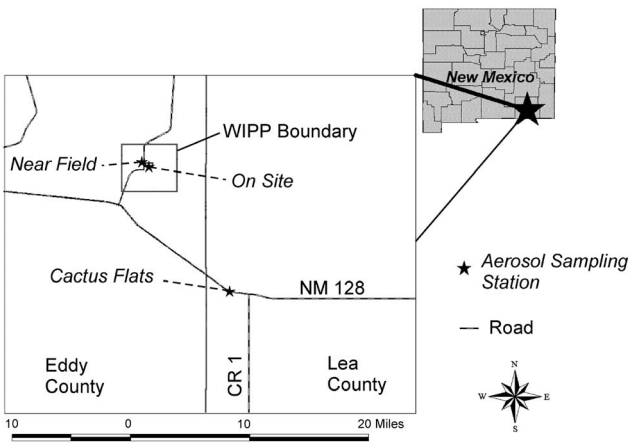

Fig. 2 Aerosol sampling locations on and near the WIPP site.

Subsequently, the actinides are separated as a group by co-precipitation on $Fe(OH)_3$. Pu isotopes are separated and purified using a two-column anion exchange resin (AG1-X8, Bio-Rad, 100–200 mesh), while TRU chromatography columns are used for the separation of Am as described previously by Thakur *et al.*, 2011.[13] The samples are then micro-coprecipitated using a Nd-carrier. A known amount of tracer (^{242}Pu, ^{243}Am or ^{232}U) is added to determine the actinide recovery in each sample. Typical chemical recoveries are in the range of "40–80%" for Pu and of "90–105%" for Am. For counting times of 72 000 min, the detection limits of $^{239+240}$Pu, ^{238}Pu and ^{241}Am are 2.6 × 10^{-9}, 3.9 × 10^{-9} and 3.1 × 10^{-9} Bq m^{-3}, respectively.

The activities of the actinides in the air samples are reported as *activity concentration* (Bq m^{-3}) and *activity density* (Bq g^{-1}). *Activity concentration* is calculated as the activity of radionuclides detected in becquerels (Bq) divided by the volume of air in cubic metres, while *activity density* is calculated as the nuclides activity divided by the aerosol mass in grams collected on the filter.

Soil sampling location and pre-treatment

Soil samples were collected at two sampling sites, including the Near Field air sampling site, which includes the set of sampling locations within the WIPP site boundary. The second site is the Cactus Flats site approximately 19 km southeast of the WIPP facility. At both sites, a grid was established with sixteen undisturbed soil sampling locations. At each sampling spot, soil was collected to a depth of approximately 2 cm. Individual sampling sites were selected on the basis of relatively flat topography, minimum surface erosion, and minimum surface disturbance by human or livestock activity. For depth profile studies, three soil samples were collected as a function of depth. The vertical profile face was scraped clean with a plastic trowel after excavation. Samples were collected by depth increment by removing a 0.25 m^2 area beginning at the profile face and working back into the undisturbed profile. A 25 cm alley was removed from around the sampled area to the depth sampled before the next depth increment was collected. Soil samples were sealed in plastic containers for transport to the lab. Additionally, nine surface soil samples were collected from near the Gnome site ground zero location to a depth of about 2.5 cm. Each soil sample was air dried, and then sieved through a 2 mm screen. Particle-size distribution was determined using the pipette method described by Gee and Bauder.[14] Specific conductivity and pH were determined on 1 : 2 (soil : deionized water) suspensions. Measurements were taken on slurry mixture for 5, 10, and 15 min. The pH values observed in each of the profiles were relatively uniform throughout, with pH values generally falling between 6.5 and 7.5. Specific conductivity values in the profiles are generally <70 μS cm^{-1} in the upper 40 cm of the soil profiles, with the exception of the immediate surface (upper 2 cm). Samples between 10 and 15 g were taken for actinide analyses. Samples for actinides were heated at 500 °C for 6 hours to combust organic material and then digested with strong acids HNO_3, HF and HCl followed by fuming with perchloric acid to remove silica. Iron hydroxides for Pu and calcium oxalate precipitation for Am were performed to separate these actinides from the soil

matrices. The chemical recoveries were in the range 60–90% for Pu and 85–105% for Am.

Quality assurance

In order to ensure the high quality of analytical results, blank, duplicate, and spiked samples were analyzed periodically according to an internal quality assurance program. Monthly checks of the background and the efficiency were performed for the measurement systems. Periodic checks of performance with an appropriate calibrated standard solution were made as part of the QA/QC program, which also included participation in the national MAPEP (DOE Mixed-Analyte Performance Evaluation Program) and NIST-NRIP (NIST-Radiochemistry Intercomparison Program) Performance Evaluation Programs. For each set of samples, reagent blank and tracer spikes were also carried through the entire separation and counting process for recovery determination and quality control.

Results and discussion

Aerosol activities

Average air concentrations of actinides in the aerosol samples during the period from 1998 to 2010 are summarized in Table 1. The average air concentrations of actinides after WIPP became operational are not statistically different from those measured prior to waste disposal operations (Table 2). Fig. 3 shows a time-series of annual $^{239+240}$Pu airborne activity concentrations observed in aerosol filters collected from OnSite, Near Field and Cactus Flats stations in the period from 1998 to the end of 2010. The annual deposition of $^{239+240}$Pu during the period from 1998 to 2010 shows no systematic interannual variation. However, monthly $^{239+240}$Pu depositions show a typical seasonal variation which is high in spring and low in summer.

A typical springtime enhanced activity of Pu was also observed in the air samples (both dry and wet deposition) collected from Japan, Hirose *et al.*,[10] and Korea.[15] The authors attributed this seasonal variation to the continental dust storms originating in the months of March to May, the so-called "Yellow Sand Event" or "Kosa" from Chinese deserts and arid regions. Furthermore, the ^{240}Pu/^{239}Pu atom ratios differed in the spring from those in the summer and autumn. Such springtime enhanced activity of radionuclides was also observed in the aerosol samples collected from Monoco,[16] Germany[17] and Sodankyla, Finland.[18] The observed seasonality in Pu activity concentration is attributed to the resuspension of contaminated soil dust plus the local precipitation to some extent. Studies conducted prior to the end of the atmospheric weapons testing showed that Pu activities varied seasonally, being highest in spring and summer because of the springtime enhanced transportation of radioactive aerosols from the stratosphere to troposphere.[19] However, with the cessation of nuclear weapon tests and considering the fact that the residence time of Pu in the atmosphere is on the order of a year,[19] the stratospheric deposition of radionuclides including Pu is no longer a dominant factor for the Pu concentration in air. Nor did the Chernobyl accident bring significant amounts of Pu to this area. Thus, resuspension is assumed to be the main source of Pu in the aerosol samples around the WIPP.

The activity concentrations of ^{241}Am in the high-volume samples closely tracked those of $^{239+240}$Pu (Fig. 3). Most notably, strong springtime peaks in ^{241}Am activity concentrations were evident in the samples from 2001 through 2002, and from 2004 through 2009. $^{239+240}$Pu and ^{241}Am are frequently detected, whereas ^{238}Pu is detected infrequently in aerosol filters, presumably because ^{238}Pu is not primarily from weapons fallout, but instead was released by the burn-up of the nuclear satellite such as SNAP-9A.[20,21] This accident was caused by the orbit failure of a satellite loaded with 17 kCi (6.3×10^{14} Bq) of ^{238}Pu. Most of the SNAP debris had been deposited predominantly (~76%) over the

Table 1 $^{239+240}$Pu, ^{238}Pu, ^{241}Am activities and the mass loadings in high-volume aerosol samples around WIPP[a]

	Statistics	Cactus Flats	Near Field	OnSite
Aerosol mass, μg m^{-3}	*N*	110	111	110
	Mean	26.64	24.32	34.38
	StdDev	13.18	9.61	14.35
^{241}Am activity concentration, Bq m^{-3}	*N*	54	51	60
	Mean	5.68×10^{-9}	4.10×10^{-9}	1.26×10^{-7}
	StdDev	4.73×10^{-9}	2.01×10^{-9}	9.43×10^{-7}
^{241}Am activity density, Bq g^{-1}	*N*	54	51	60
	Mean	2.21×10^{-4}	1.66×10^{-4}	5.47×10^{-3}
	StdDev	1.78×10^{-4}	7.21×10^{-5}	4.14×10^{-2}
^{238}Pu activity concentration, Bq m^{-3}	*N*	9	2	10
	Mean	6.13×10^{-9}	1.20×10^{-9}	2.98×10^{-9}
	StdDev	1.14×10^{-8}	4.61×10^{-10}	1.47×10^{-9}
^{238}Pu activity density, Bq g^{-1}	*N*	9	2	10
	Mean	2.69×10^{-4}	3.76×10^{-5}	8.24×10^{-5}
	StdDev	5.45×10^{-4}	5.14×10^{-6}	3.30×10^{-5}
$^{239+240}$Pu activity concentration, Bq m^{-3}	*N*	89	100	97
	Mean	1.53×10^{-8}	1.22×10^{-8}	1.21×10^{-8}
	StdDev	1.12×10^{-8}	8.52×10^{-9}	7.68×10^{-9}
$^{239+240}$Pu activity density, Bq g^{-1}	*N*	89	100	97
	Mean	5.22×10^{-4}	4.80×10^{-4}	3.42×10^{-4}
	StdDev	2.01×10^{-4}	2.22×10^{-4}	1.89×10^{-4}

[a] *N* = number of samples with masses or activities >MDC.

Table 2 The background concentration of actinides (Bq m^{-3}) in New Mexico air

	EEG[a]	LANL[b]	EPA[c]	WID[d]
^{241}Am	$2.3 \times 10^{-8} \pm 3.6 \times 10^{-8}$	$8.9 \times 10^{-8} \pm 5.9 \times 10^{-8}$	N/A	$2.2 \times 10^{-7} \pm 5.3 \times 10^{-7}$
$^{239+240}$Pu	$1.7 \times 10^{-8} \pm 1.6 \times 10^{-8}$	$2.2 \times 10^{-8} \pm 4.8 \times 10^{-8}$	$5.9 \times 10^{-9} \pm 8.5 \times 10^{-9}$	$3.7 \times 10^{-7} \pm 2.7 \times 10^{-7}$
^{238}Pu	$-9.0 \times 10^{-9} \pm 2.6 \times 10^{-8}$	$5.6 \times 10^{-9} \pm 2.2 \times 10^{-8}$	$1.3 \times 10^{-8} \pm 1.4 \times 10^{-8}$	$3.7 \times 10^{-7} \pm 2.7 \times 10^{-7}$

[a] Average concentrations near the WIPP site during 1998. [b] Average concentrations in air samples collected from Santa Fe, Espanola and Pojoaque, New Mexico during 1996. [c] Average concentrations in air samples collected from Santa Fe, during 1996. [d] Average concentrations near the WIPP site collected by WID during 1996–97 and USDOE, WIPP 1998.

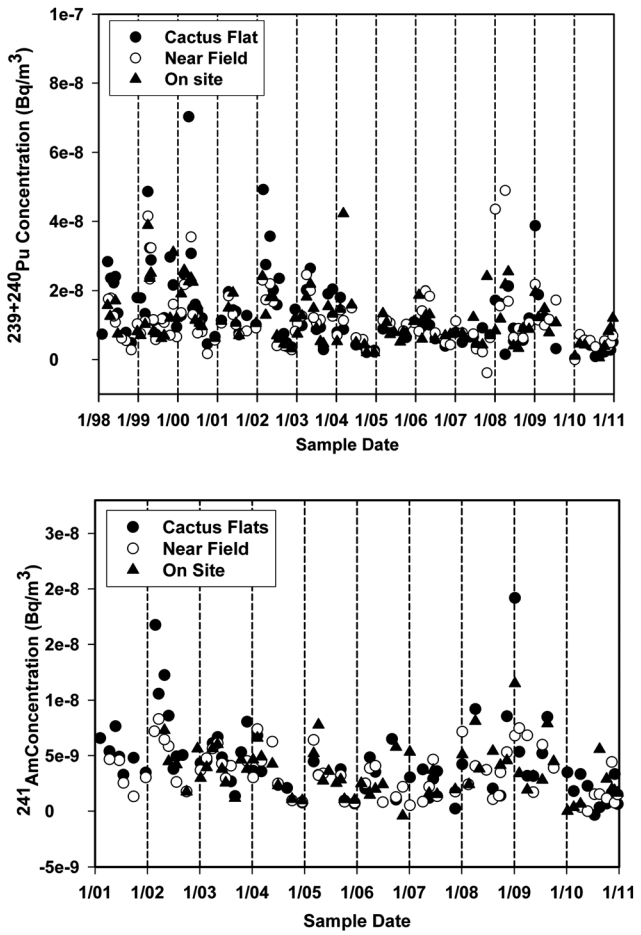

Fig. 3 Temporal patterns in $^{239+240}$Pu (top) and ^{241}Am (bottom) atmospheric activity concentrations at three stations in the vicinity of the WIPP site.

southern hemisphere, with only 24% over the northern hemisphere. Approximately the same, but reversed, was the distribution of the deposited radionuclides due to atmospheric weapon tests. Because of this, the ^{238}Pu/$^{239+240}$Pu ratio for northern hemisphere was increased after 1964 (0.036 for the northern hemisphere and 0.142 for the southern hemisphere),[20] but indicating that about 5% of total ^{238}Pu was still in the stratosphere. In cases where ^{238}Pu was detected, its activity tended to increase with $^{239+240}$Pu, indicating that ^{238}Pu, too, may be recycled between the earth and atmosphere on contaminated soils. Table 3 compares the Pu results in the aerosol samples collected in the vicinity of the WIPP sites with those collected around the world.

Ambient air particulate activity concentrations of $^{239+240}$Pu range from 2.1–42.3 nBq m^{-3} at OnSite, 1.7–41.6 nBq m^{-3} at Near Field, 2.1–70.3 nBq m^{-3} at Cactus Flats in broad agreement with previously published data (10 to 100 nBq m^{-3}) in the continental US (Kirchner *et al.*, 2002).[22] But it is clearly higher than the concentration range of 0.4 to 2.9 nBq m^{-3} found in northern Germany[17] and the 1.0 to 22.2 nBq m^{-3} range determined in air samples collected from Lublin, Poland during 1998–1999.[23] In contrast, the $^{239+240}$Pu activity concentrations in the WIPP aerosol samples are lower than those at Kurchatov (14 to 2500 nBq m^{-3}) and at several villages just outside the Semipalatinsk nuclear test site (STS),[24] at Anmyeondo, Korea (<4.0 to 580 nBq m^{-3})[15] and in Lithuania (1 to 500 nBq m^{-3}).[25] Furthermore, aerosol mass loadings follow a seasonal pattern similar to that observed for $^{239+240}$Pu. Activity density, used as an index to elucidate the contribution of resuspended $^{239+240}$Pu on the aerosol filters,[10] ranges from 0.13–0.41 mBq g^{-1} at OnSite, 0.19–0.77 mBq g^{-1} at Near Field, and 0.12–0.59 mBq g^{-1} at Cactus Flats.

The highest levels of activity both in terms of activity concentrations and activity densities occurred not at the OnSite station, which is where one would expect any emission from the WIPP to be most evident, but rather at Cactus Flats, the reference station farthest from WIPP. The Pu activity concentration follows the order: Cactus Flats > Near Field > OnSite. In contrast to actinide data, the aerosol mass loadings follow the trend: OnSite (34.4 ± 14.3 μg m^{-3}) > Cactus Flat (26.2 ± 13.2 μg m^{-3}) > Near Field (24.3 ± 9.6 μg m^{-3}). Furthermore, aerosol mass loading shows a linear relationship with the $^{239+240}$Pu activity concentrations for the Near Field and Cactus Flats stations. However, such linearity is not observed for the OnSite station (Fig. 4). The mass loading is usually high with low $^{239+240}$Pu activity concentration at the OnSite station. This suggests that operations at the WIPP such as underground salt mining, construction or road dust may have generated some aerosols that contributed to the mass loadings but contain less $^{239+2340}$Pu than ambient aerosols typically do.

Time series plots, not presented here, for Al, the elemental indicator for mineral dust, also show the same springtime peaks as the actinides indicating temporal variability in $^{239+240}$Pu and ^{241}Am synchronous with the seasonal cycle of atmospheric dust. The Al mass loadings and the $^{239+240}$Pu activity concentrations are highly correlated ($r^2 = 0.57$), indicating that Pu is transported with mineral dust. The WIPP soils study of Kirchner *et al.*[22] also showed that the activities of radionuclides in soils were strongly

Table 3 Comparison of $^{239+240}$Pu concentration in the aerosol samples collected around the WIPP site with those collected around the world

	Location	Activity (nBq m^{-3})	Year	Ref.
$^{239+240}$Pu	OnSite, WIPP	2.1–42.3	1998–2010	This work
	Near Field, WIPP	1.7–41.6	1998–2010	This work
	Cactus Flats, WIPP	2.1–70.3	1998–2010	This work
	Continental US	10–100		53
	Nevada Test Site, USA	4×10^6		21
	Northern Germany	0.4–2.9	1990	17
	Neuherberg, Munich	2–6	1987–1991	3
	Lublin, Poland	1.0–22.2	1998–1999	23
	Kurchatov, Semipalatinsk Nuclear Test Site, Kazakhstan	14–2500	2000–2001	24
	Astana, Kazakhstan	5–131	2001	24
	Dolon, Kazakhstan	7400–37 000	1991–1992	24
	Anmyeondo, Korea	<4.0–580		15
	Lithuania	1–500		25
	Prague, Czech	0.5–5.1	1997–2003	54
	Lapland, Finland	400–95 000	1963	18
	Palomares, Almeria, Spain	5000–55 000		55
	Seville, Spain	14.7–18.8		56
	Madrid, Spain	13		57

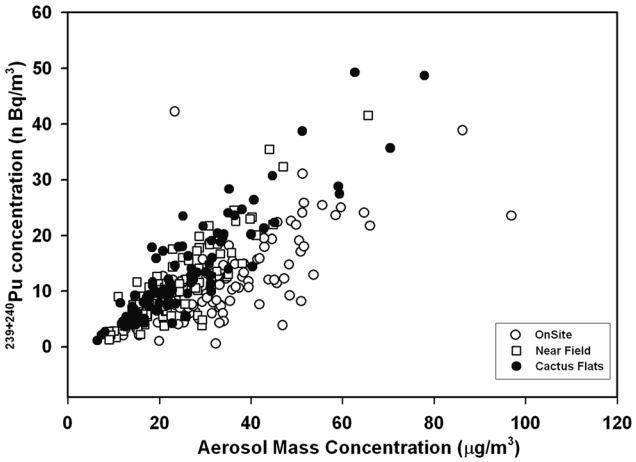

Fig. 4 Aerosol mass *vs.* $^{239+240}$Pu activity concentrations in high volume aerosol samples at the three stations near the WIPP site.

correlated with the concentrations of Al and Pb, and with the percentages of sand, silt, and clay in the soil.

The suspension of soil particles typically exhibits a threshold with respect to wind velocity. Thresholds typically range from 6 to 13 m s^{-1} at 0.3 m and, they are related to the force required to cause the saltation of large soil particles. The regression analysis ($r^2 = 0.5$ for threshold velocity >8 m s^{-1})[5] demonstrates that activity of $^{239+240}$Pu increases as a function of wind speed, supporting the theory that the activities of $^{239+240}$Pu are controlled by resuspension. Further, threshold wind velocity studies for sand movement in southeastern New Mexico demonstrate that the dispersal of contaminated soils from the Gnome site is likely to occur only when winds are greater than ~10 to 10.5 m s^{-1}.[26] This occurs generally in the months of April through May when winds are strong, precipitation is low, and the air is drier. In addition, the results show that saltation activity is favored at certain times of the day, especially from noon to mid-afternoon.

Activity ratios in aerosols

A strong correlation between aerosol ^{241}Am and $^{239+240}$Pu activity concentrations exists ($r^2 = 0.75$, 0.72, and 0.76 respectively, for OnSite, Near Field and Cactus Flats stations) even though

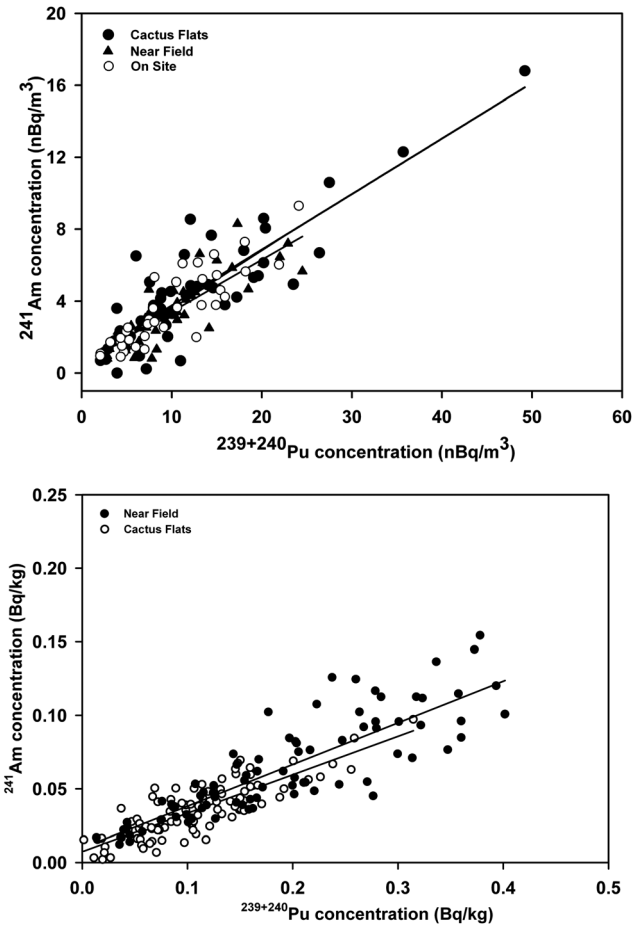

Fig. 5 Correlation between ^{241}Am and $^{239+240}$Pu in aerosol (top) and soil (bottom) samples.

neither ^{239}Pu nor ^{240}Pu are the immediate progeny of ^{241}Am (Fig. 5). Furthermore, the relative activities of ^{241}Am and $^{239+240}$Pu were similar at all aerosol stations. The mean activity ratios of ^{241}Am/$^{239+240}$Pu (as of Jan 2010) at three aerosol stations were found to be 0.37 ± 0.05 (OnSite), 0.37 ± 0.02 (Near Field), and 0.41 ± 0.04 (Cactus Flats). These values are in agreement with the mean activity ratio of these radionuclides, originating from the atmospheric nuclear tests in soils and sediments reported in different studies: 0.32, 0.35,[27] 0.30,[28] 0.34, 0.37, 0.39, 0.42,[29] 0.43.[30] Taking into account the contribution of ^{241}Am from ^{241}Pu decay, the observed ^{241}Am/$^{239+240}$Pu activity ratio again strongly indicates global fallout origin.

The ^{238}Pu/$^{239+240}$Pu activity ratios in aerosol filters collected near WIPP varied from 0.069 ± 0.02 to 0.12 ± 0.04. Because of the normally low levels of ^{238}Pu in the environment, the measured ratio ^{238}Pu/$^{239+240}$Pu has high uncertainty values of at least 15–25%. These values are clearly higher (>2×) than the value reported for global fallout in different studies: Bunzl and Kracke[31] measured 0.031 (Jan 2010) for soil in South Germany in 1988, Holgye and Filagas, 0.029 (Jan 2010) for soil in Czechoslovakia in 1990,[32] Holgye *et al.*,[33] 0.039 (0.025–0.059) (Jan 2010) near nuclear research center, Prague (Czech Republic) in 2004, Breban *et al.*,[34] 0.044–0.054 (Jan 2010) for the Carpathian Mountain (Romania) in 1996, Mietelski *et al.*,[35] 0.042, for the Tatra National Park (Poland), 0.034 in the Marche region of central Italy,[27] 0.033 (0.025–0.057) (Jan 2010) for soil in Korea during 1992–1994,[36] and 0.034 for the Irish soil.[37] At present, we do not have logical explanations for these high ^{238}Pu/$^{239+240}$Pu ratios measured in aerosol samples in the vicinity of the WIPP site. However, it can be assumed that dust released from top soil during strong wind seasons might be the reason for the increased Pu ratio. Additional studies are needed to obtain a better understanding of the behavior of Pu in the WIPP environment. The high ^{238}Pu/$^{239+240}$Pu ratio as in the present investigation is also reported in aerosol samples taken from Krakow by Mietelski *et al.*,[38] while unusually high ^{238}Pu/$^{239+240}$Pu activity ratios of 0.36 (Jan 2010) and 0.041 (Jan 2010) were found in three quarterly aerosol samples collected from Germany.[17] Similarly, the high ^{238}Pu/$^{239+240}$Pu activity ratio of 0.42 (Jan 2010) was reported in aerosol samples collected from Lublin, Poland in 1999.[23]

Soil activities

The activity concentration of ^{137}Cs in the Near Field surface soil ranged from 0.50 to 5.12 Bq kg^{-1}, with a mean value of 3.29 Bq kg^{-1}. The $^{239+240}$Pu concentrations in the Near Field ranged from 0.02 to 0.24 Bq kg^{-1}, with a mean value of 0.01 Bq kg^{-1}, while that for ^{241}Am ranged from 0.001 to 0.07 Bq kg^{-1}, with a mean value of 0.004 Bq kg^{-1}. The concentrations of these nuclides are comparable to historical data recorded in 1997 prior to the arrival of TRU wastes in the WIPP (Table 4). Surface soils were not collected from Cactus Flats in 2007 through 2009. However those collected in 1998 to 2006 show activity concentrations of $^{239+240}$Pu ranging from 0.01 to 0.51 Bq kg^{-1}, while that of ^{241}Am varied from 0.10 to 0.26 Bq kg^{-1}. The ^{137}Cs concentration ranged from 0.69 to 14.83 Bq kg^{-1}. The concentration of ^{137}Cs in the surface soil at Cactus Flats is approximately three times higher than that of the surface soil at Near Field, while concentrations of $^{239+240}$Pu and ^{241}Am are *ca.* 2–3 times higher. However, there is no apparent difference between the radionuclides concentration in soil collected before and after WIPP started receiving TRU waste. On the other hand, all of the Gnome soil samples show elevated concentrations of ^{137}Cs as compared to the soils around the WIPP site. The maximum observed concentration of ^{137}Cs for the Gnome samples, 2890 Bq kg^{-1} (45.9 to 2890 Bq kg^{-1}), was more than 100 times larger than the largest concentration seen previously in the WIPP soils, while those of $^{239+240}$Pu, ^{238}Pu, and ^{241}Am were in the range 0.007–1550 Bq kg^{-1}, 0.016–219 Bq kg^{-1} and 0.004–346 Bq kg^{-1}, respectively with an overall mean of 149 Bq kg^{-1}, 28.8 Bq kg^{-1} and 36.1 Bq kg^{-1}.[39] Kenney *et al.*[40] reported activity concentrations of $^{239+240}$Pu in the range 4.4 to 48 000 Bq kg^{-1}, ^{238}Pu 0.81 to 6800 Bq kg^{-1} and ^{241}Am 0.59 to 7600 Bq kg^{-1} for the Gnome soil in 1995. Concentrations of ^{238}Pu, $^{239+240}$Pu, and ^{241}Am for one of the Gnome samples were more than 1000 times greater than the maximum concentrations observed in WIPP soils. However, the remaining samples showed at most only moderately higher actinide activity concentrations and many of these samples fell within the range of concentrations previously observed in WIPP soils. The wide range in depositions can probably be because of the spotty and inconsistent cleanup of the Gnome site.[12] From 1968–1969, a clean-up program was conducted with the goal of removing all materials exhibiting radiation levels greater than 0.1 mR h^{-1} (beta plus gamma) as measured with a 30 mg cm^{-2} window Geiger–Müller survey meter.[12] The contaminated soil was either disposed of in the Gnome shaft and drift tunnels or buried on site. All surface facilities were removed and the boreholes plugged except for those used for hydrological monitoring. In 1977, a second more extensive clean-up program was initiated with a goal to remove soil having alpha plus beta radiation exceeding 0.74 Bq g^{-1} (20 pCi g^{-1}).[12] Thus, in spite of the earlier remediation efforts at the Gnome site, there are some areas where levels of radionuclide contamination remain elevated relative to the levels in the testing area. These contaminated soils are a potential source of airborne contamination when collecting WIPP air particulate samples. A very good correlation between

Table 4 The background concentrations of ^{137}Cs, $^{239+240}$Pu and ^{241}Am (Bq kg^{-1}) in surface soil around the WIPP site

Location	Year	^{137}Cs	$^{239+240}$Pu	^{241}Am
WIPP OnSite	1998	1.96–11.65	0.074–1.41	0.037–0.259
WIPP vicinity, Near Field	1998	0.29–5.92	0.015–0.185	0.0148–0.148
WIPP vicinity, Cactus Flats	1998	0.70–14.98	0.026–0.518	0.022–0.026
WIPP vicinity	1995	0–7.40	0–0.74	—
Gnome	1995	2.59–3086	4.4–48 000	0.4–7600
Gnome	2002	45.9–2980	0.07–1550	0.04–3460
Distant locations	1982–1987	6.45–47.25	0.13–6.98	—

$^{239+240}$Pu and ^{241}Am concentrations was also observed for both Near Field ($r^2 = 0.72$) and Cactus Flats ($r^2 = 0.68$) soil samples, which suggests identical origin (Fig. 5).

The range of $^{239+240}$Pu in the Near Field soil is lower than background concentrations found at Hueston Woods and Urbana, Ohio (0.7–1.0 Bq kg^{-1}),[41] and those found between Ft. Collins and Colorado Springs, Colorado (0.6 to 1.7 Bq kg^{-1}).[22] The $^{239+240}$Pu concentrations in the WIPP vicinity are also lower than those found in the background soil at Rocky Flats, Colorado, control (0.4–2.0 Bq kg^{-1}) and at Hanford, Washington (0.3 Bq kg^{-1}).[42] The deposition of radioactive fallout from weapons testing is known to vary with latitude, being highest in middle latitudes of the northern hemisphere.[2]

Vertical distribution of ^{137}Cs, ^{241}Am and $^{239+240}$Pu

The vertical distribution of ^{137}Cs, $^{239+240}$Pu, and ^{241}Am activities as a function of depth in soil profiles is utilized to evaluate the dynamics of transport of these radionuclides over the last four decades in a desert environment. ^{137}Cs concentrations in soil were detectable down to a depth of 20 cm. From the results obtained, it can be calculated that approximately 60% of ^{137}Cs was retained in the top 3 cm of soil and 86% was within the top 6 cm. An exponential decrease with depth was observed for ^{137}Cs ($r^2 = 0.93$) for all soil profiles (Fig. 6). This is in agreement with the soil depth profile studies at Queen City summit, 55 km north of the Nevada Test Site (NTS), at Searchlight, 160 km southeast of the NTS, by Turner *et al.*[42] and at Rocky Flats, Hulse *et al.*[43] There is no indication that radionuclides have mobilized to any substantial degree within these profiles. It is possible that the increase in ^{137}Cs activities with a slight increase in depth could be the result of leaching. However, it is also possible that the immediate surface is characteristic of a "mixing zone" in which the sandy surface could be depositional, erosional, or both.

The distribution of $^{239+240}$Pu was more irregular, in terms of overall trends, than those for ^{137}Cs. The highest $^{239+240}$Pu concentration was found at surface layers and at the layer from 3–4 cm depth; it is also detectable down to 10 cm (Fig. 6). The results show an exponential decrease of the activity with depth, in the same manner as was observed by Mitchell *et al.*[37] in soil samples collected from Ireland, Ibrahim *et al.*[44] in Rocky Flats soil, Lee *et al.*[45] in Korean soil, and Yamamoto *et al.*[46] in soil samples obtained from Japan. For all profiles, about 70% of the Pu is contained in the first 5 cm of soil depth; and are at, or below, minimum detectable concentrations (MDC) below 10 cm of the surface soil, which is consistent with the depth distribution of Pu reported by Bunzl and Kracke[31] and Komosa.[47] The highest concentrations of ^{241}Am in all three profiles also occurred in 2 and 3 cm depth. However, because there are relatively few data points, it is impossible to discern a trend for ^{241}Am.

Although chemically quite different, ^{137}Cs is expected to behave similar to $^{239+240}$Pu in dry soils because it adheres tightly to cation-exchange surfaces. ^{137}Cs from fallout has been frequently used to characterize soil particle transport. Cesium can adsorb to organic matter as well as soil mineral particles. However, cesium adsorption on clay minerals is strongly specific, whereas adsorption on organic matter is considered to be non-specific. Therefore, even when organic matter is responsible for a large proportion of soil cation-exchange capacity, trace amounts of radiocesium will be preferentially adsorbed on clay minerals. Nevertheless, organic matter can play a role in the mobility of cesium in the soil. Very good correlations between concentration of ^{137}Cs and $^{239+240}$Pu ($p = 0.0009$) were observed, although the r^2 value was only 0.35 because the slope was close to zero (Fig. 7). Profile 1 had two relatively large deviations from the regression

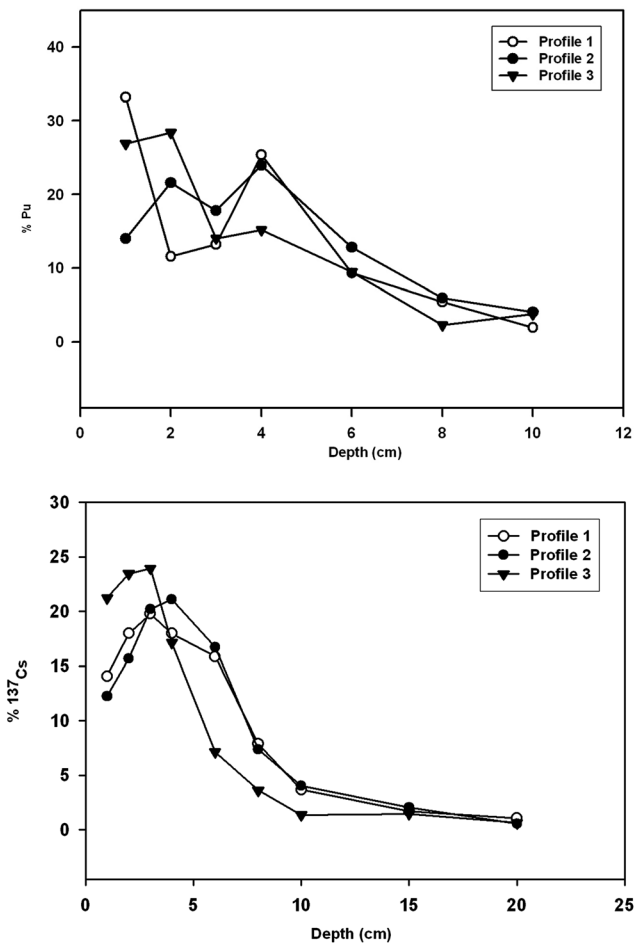

Fig. 6 Distribution of $^{239+240}$Pu (top) and ^{137}Cs (bottom) as a function of depth.

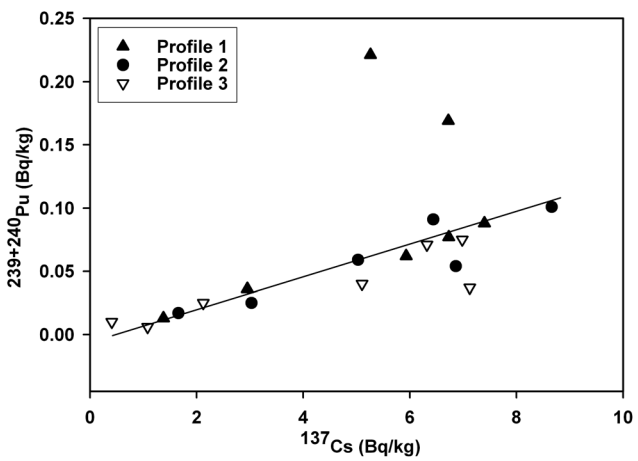

Fig. 7 Correlation between $^{239+240}$Pu and ^{137}Cs in the soil profile.

line. This implies that both ^{137}Cs and $^{239+240}$Pu move at similar rates and are probably transported by similar mechanisms in soil around WIPP. WIPP soils are dominated by sand and because Cs and Pu have affinities for binding to clay particles, the distribution of clay could be a factor affecting the distribution of Cs and Pu in the soil. The clay percentages in the WIPP soil ranged from 2 to 5%. However, there appears to be no correlation between the percentage of clay in a soil layer and the concentration of ^{137}Cs in that layer. The pH, another representative soil characteristic, had a weak correlation with the concentration of ^{137}Cs in the soil, which is consistent with the previous report.[48] Based on *in situ* soil profiles and on the soil column experiments, it is apparent that leaching and colloidal transport are not major factors in affecting the vertical movement of ^{137}Cs or $^{239+240}$Pu in the soils found in the vicinity of WIPP. The lateral movement of soil by wind erosion is, by far, more responsible for the redistribution of the radionuclides in this ecosystem.

Activity ratios in soil

Activity ratios between ^{137}Cs/$^{239+240}$Pu, ^{238}Pu/$^{239+240}$Pu and ^{241}Am/$^{239+240}$Pu were calculated in order to understand the source of these radionuclides in soils. The average value of ^{137}Cs/$^{239+240}$Pu for northern hemisphere fallout is 36 ± 4 (as of July 1995 for the decay of ^{137}Cs, half-life 30.5 years).[42] Additionally, values of 33 and 30 (as of 1 July 1995) are reported for radionuclide fallout in glaciers in Greenland and Antarctica.[42] Perturbation in this ratio could result if soil chemistry favored the downwind migration of one of these radionuclides or possible contamination by sources other than fallout. For example Price[49] measured ^{137}Cs/$^{239+240}$Pu ratios of 4.7 and 3.6 in soil samples collected in the vicinity of the Hanford reprocessing plant compared to a mean ratio of 32 (decay corrected to July 1998) found at a site 25 km from the reprocessing plant; this indicates possible contamination from the Hanford plant. Similarly, low ^{137}Cs/$^{239+240}$Pu ratio in the soil samples from Searchlight (12 to 28) and Queen City (0.8 to 9) is attributed to the contribution of contaminants from NTS.[42] The mean ratio of ^{137}Cs/$^{239+240}$Pu across all sampling grids around WIPP was found to be 30 ± 5. There was no significant difference between the mean ratios found for the Cactus Flats (29 ± 4) and Near Field soils (30 ± 7). Correcting these two ratios for decay to July 1, 1999 yields ratios of 33 ± 4 and 34 ± 4, respectively. Although our ratio is somewhat lower than previously reported data for global fallout, it falls within the reported uncertainty associated with those ratios. This agreement suggests that the WIPP soils have received their Pu and Cs from world-wide fallout from the nuclear weapons testing and are typical "background" soils. In contrast, the mean ^{137}Cs/$^{239+240}$Pu ratios for the Gnome soil (3591 ± 1635) are significantly higher than the corresponding mean found in the WIPP soils. However, variability is very great in the Gnome sample which is not surprising given the inconsistent and discrete clean-up of the site.

The ^{238}Pu/$^{239+240}$Pu activity ratios in the WIPP soil ranged from 0.06 to 0.19 with a mean value of 0.14 ± 0.05. This ratio is comparable to a mean observed ratio of 0.13 ± 0.03 (ref. 39) and 0.16 ± 0.02 reported previously for the Gnome soils.[40]

The mean activity ratio of ^{241}Am/$^{239+240}$Pu in WIPP soils was 0.36 ± 0.03 (Near Field) and 0.39 ± 0.04 (Cactus Flats) which is indicative of global fallout origin. The mean ratio in the WIPP soil is comparable to the mean observed ratio of 0.35 ± 0.03 for Gnome soils.

^{240}Pu/^{239}Pu atom ratio in soils

The ^{240}Pu/^{239}Pu atom ratio in the WIPP soil ranged from 0.193–0.146 with a mean value of 0.175 ± 0.005, while the corresponding ratio in the Gnome soils ranged from 0.07–0.168 with a mean value of 0.114 ± 0.013. The errors shown are one standard deviation obtained from AMS (Accelerator Mass Spectrometry) measurements. Krey *et al.*[50] reported the atomic ratio of ^{240}Pu/^{239}Pu in global fallout to be 0.176 ± 0.014 ranging from 0.12 to 0.21 around the world, based on a world-wide program of sampling conducted at 21 sites in 1970–1971 between 30 and 60° N, established following a worldwide survey of terrestrial soils in 1970–1971. The exceptions in the northern hemisphere are associated with soils collected in the southwestern United States, where fallout from the NTS was also deposited.[51] The ^{240}Pu/^{239}Pu atom ratio produced in a nuclear test is a function of the design and yield of the device being tested. The fallout produced by high yield tests tends to have higher ^{240}Pu/^{239}Pu atom ratios than the fallout produced by lower yield nuclear tests *i.e.*, the fallout from Nagasaki, Nevada Test Site and Semipalatinsk Test Site is generally characterized by lower ^{240}Pu/^{239}Pu ratios (0.03–0.08) with average values of 0.042, 0.035 and 0.036, respectively, whereas elevated ratios (0.21–0.36) have been measured in soils from Bikini atoll.[52]

Since ratios were determined on bulk soils, the high variability of ^{240}Pu/^{239}Pu atom ratios in the Gnome samples could be due to measurements error on low activity samples or there is non-uniform distribution of radioactive particles within a sample. The Gnome sample having the greatest activity concentration (743 Bq kg^{-1}) had a ^{240}Pu/^{239}Pu atom ratio of 0.074 ± 0.003. This value is probably a good estimate of the ^{240}Pu/^{239}Pu atom ratio of the Gnome fallout. The Gnome ^{240}Pu/^{239}Pu ratio fell within the range of those observed at the NTS (0.00015 to 0.082). Three of the Gnome samples had ^{240}Pu/^{239}Pu atom ratios within the range of the ratios measured for the WIPP samples. The activity concentrations of these three samples also fall within the range of the WIPP samples. Two of these samples were collected along the line of the original fallout plume and one from the side of a road used to haul contaminated materials during cleanup of the site. Given the cleanup of the site, it would not be surprising if the plutonium in these samples came from global fallout. Although these samples had relatively low activity concentrations of $^{239+240}$Pu, activity concentration alone is not a good predictor of ^{240}Pu/^{239}Pu atom ratio. Three of the remaining Gnome samples fall within or below the range of activity concentrations observed in the WIPP samples. Because of the large variability in ^{137}Cs/$^{239+240}$Pu ratios, the ^{240}Pu/^{239}Pu ratio represents the most reliable "fingerprint" for identifying Gnome-contaminated soil. Literature values of ^{238}Pu/$^{239+240}$Pu and ^{137}Cs/$^{239+240}$Pu activity ratios as well as ^{240}Pu/^{239}Pu atom ratios from different sources which can be used as an indicator for identifying the source of these radionuclides are summarized in Tables 5 and 6. The activity ratios of ^{238}Pu/$^{239+240}$Pu, ^{137}Cs/$^{239+240}$Pu, and ^{241}Am/$^{239+240}$Pu vary according to the source, so they can be utilized to identify the origin of the Pu from different sources released into the environment.

Table 5 $^{240}Pu/^{239}Pu$ atom ratios from different sources

Source	Remark	$^{240}Pu/^{239}Pu$	Ref.
Global fallout, northern hemisphere	Soil	0.176 ± 0.014 (0.12 to 0.21)	50
Global fallout, southern hemisphere	Soil	0.142	21
Northern New Mexico	Environmental	0.169 ± 0.012	58
Gnome	Soil	0.114 ± 0.013	39
WIPP	Soil	0.175 ± 0.005	39
Chernobyl accident		0.38–0.52	59
Nevada Test Site	Soil	0.03–0.075	10
Nagasaki, 1979	Soil	0.042 ± 0.014	10
Semipalatinsk Test Site	Soil	0.036	10
Thule hydrogen bomb debris	Hot particles	0.0551 ± 0.0008	60
Thule	Sediments	0.058 ± 0.008	60
Palomares, Spain	Sediments	0.056 ± 0.003	61
Mayak Nuclear Installation	Soil/sediments	0.1–0.3	59
Sellafield		0.05–0.25	59
Pressurized heavy water reactor	—	0.41[a]	62
Advanced gas cooled reactor	—	0.57[a]	62
Boiling water reactor	—	0.40[a]	62
Pressurized water reactor	—	0.43[a]	62
BOMARC Missile Site[b]	Soil	0.0559–0.0594	63
Trinity Test Site, 1973–74	Soil and air	0.02	58
Rocky Flats plant	Sediment/soil	0.06	58
Mururoa Atoll	Ocean	0.043 ± 0.008	52
	Sediment	0.036–0.056	
Bikini Atoll	Soil	0.30	52
Marshall Island	Soil	0.06–0.31	52
Lithuania	Filter	0.03–0.19	25

[a] After fuel burn-up. [b] Boeing Michigan Aeronautical Research Center.

The mean activity ratio of $^{238}Pu/^{239+240}Pu$ and $^{241}Am/^{239+240}Pu$ in aerosol samples is similar to those found in soils around the WIPP. Although the particle distributions of the soils and aerosols are quite different and that composition likely varies with particle size, the similarity of $^{238}Pu/^{239+240}Pu$ and $^{241}Am/^{239+240}Pu$ activity ratios in the two studies does support

Table 6 $^{238}Pu/^{239+240}Pu$ and $^{137}Cs/^{239+240}Pu$ activity ratios from different sources

Source	$^{238}Pu/^{239+240}Pu$	$^{137}Cs/^{239+240}Pu$	Ref.
Global fallout, northern hemisphere	0.03	36 ± 4	21
Global fallout, southern hemisphere	0.20	—	20
WIPP soil	0.14 ± 0.05	30 ± 5	39
La Hague 66-82	0.50	—	64
Gnome soil	0.16 ± 0.02	3591 ± 1635	39
Chernobyl, 1986, 1992	0.45	—	64
	0.50	—	
Shellafield 93	0.29	—	64
Chernobyl, calculated, 1986	0.33	—	64
Chernobyl contaminated soil, 1996	0.41 ± 0.05	—	64
Russian soil, 1986	0.37 ± 0.05	—	64
	0.27 ± 0.05	—	
	0.30 ± 0.03	—	
Thule hydrogen bomb debris, 2001	0.0161 ± 0.0005	—	60
Surface air, Sodankyla, 1963	0.045	—	25
Northwestern Mediterranean sea, after decommissioning of Marcoule reprocessing plant	0.06–0.24	—	65
Marcoule Reprocessing Plant	0.31 ± 0.09	—	65
Weapons grade plutonium, Mururoa Atoll, 1997	0.0054 ± 0.002	—	66
Soil contaminated with weapons grade plutonium, Fangataufa Atoll, 1997	0.018 ± 0.003	—	66
Soil contaminated with weapons grade plutonium, Palomares, Spain, 1997	0.31 ± 0.09	—	66
BOMARC Missile Site	0.017–0.033	—	63
NTS only fallout, 1995	—	21–6	42
Queen City summit, Nevada	—	12–78	42
Searchlight, Nevada	—	9–35	42
Korean soil, 1994	0.038 ± 0.008	31–76	36
Northern Bulgaria soil	0.061 ± 0.022	151 ± 97	67
Semipalatinsk Test Site	0.0145–0.447	—	68

a connection between contaminated soil and aerosol. The mean $^{240}Pu/^{239}Pu$ atom-ratio observed in the WIPP samples (0.175) is consistent with the source being largely global fallout. Thus it is likely that most of the Pu in the WIPP environment comes from atmospheric resuspension of soil particles with Pu deposited from global fallout. The variation in activity ratio as well as concentration is likely because of variations in the resuspension conditions and atmospheric transport and dispersion conditions.

Conclusion

The source and level of plutonium in and around the WIPP environment prior to the arrival of the TRU waste and after it became operational were compared to assess if there is any evidence of an increase in radionuclide activity concentrations in the region that could be attributed to releases from WIPP. After twelve years of continuous operations, there is no evidence of increases in radiological contaminants in the region that could be attributed to releases from WIPP. Fallout in the vicinity of the WIPP is a mixture of global and local fallout from the above ground testing at the Nevada Test Site. Although transport of contaminants from the Gnome site to the WIPP remains a possibility during high wind seasons, the activity and the atomic ratio measurements indicate that the deposited Pu in the aerosol and soil samples mainly results from global fallout, with possible minor contamination from Gnome site or NTS. Resuspension of soil particles which are contaminated from weapons fallout continues to be considered the predominant source of plutonium in the environment surrounding the WIPP area.

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