



# Sources and distribution of $^{241}\text{Am}$ in the vicinity of a deep geologic repository

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Received: 17 September 2018 / Accepted: 6 November 2018 / Published online: 21 November 2018

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## Abstract

The detection, distribution, and long-term behavior of  $^{241}\text{Am}$  in the terrestrial environment at the Waste Isolation Pilot Plant (WIPP) site were assessed using historical data from an independent monitoring program conducted by the Carlsbad Environmental Monitoring & Research Center (CEMRC), and its predecessor organization the Environmental Evaluation Group (EEG). An analysis of historical data indicates frequent detections of trace levels of  $^{241}\text{Am}$  in the WIPP environment. Positive detections and peaks in  $^{241}\text{Am}$  concentrations in ambient air samples generally occur during the March to June timeframe, which is when strong and gusty winds in the area frequently give rise to blowing dust. A study of long-term measurements of  $^{241}\text{Am}$  in the WIPP environment suggest that the resuspension of previously contaminated soils is likely the primary source of americium in the ambient air samples from WIPP and its vicinity. Furthermore, the  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratio in aerosols and soils was reasonably consistent from year to year and was in agreement with the global fallout ratios. Higher than normal activity concentrations of  $^{241}\text{Am}$  and  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratios were measured in aerosol samples during 2014 as a result of February 14, 2014 radiation release event from the WIPP underground. However, after a brief spike, the activity concentrations of  $^{241}\text{Am}$  have returned to the normal background levels. The long-term monitoring data suggest there is no persistent contamination and no lasting increase in radiological contaminants in the region that can be considered significant by any health-based standard.

**Keywords** Americium · WIPP site · Aerosol · Resuspension · Soil · Depth distribution

## Introduction

Americium (Am) is an anthropogenic,  $\alpha$ -emitting radionuclide produced in small quantities in nuclear reactors and by the decay of  $^{241}\text{Pu}$ . Its occurrence in the environment can be attributed to three sources: fallout from nuclear weapons testing, releases from nuclear reactors and reprocessing plants, and production and disposal of smoke detectors by producers and consumers. The americium isotopes range from  $^{232}\text{Am}$  to  $^{247}\text{Am}$ , of which  $^{241}\text{Am}$  is the most important isotope found in

the environment. It is also the most prevalent isotope of americium in nuclear waste and because of the low penetration of  $\alpha$ -radiation,  $^{241}\text{Am}$  only poses a health risk when ingested or inhaled (UNSCEAR, Report 1982).  $^{241}\text{Am}$  also emits 59.54 keV gamma radiation with an intensity of 36%. However, due to its low energy, the health risk from gamma is significantly lower than that from alpha emissions of  $^{241}\text{Am}$ . Not only is the half-life ( $t_{1/2}$ ) of 432.2 years much longer than that of its  $^{241}\text{Pu}$  parent ( $t_{1/2} = 14.3$  years), but  $^{241}\text{Am}$  is much more radiotoxic and mobile. The  $^{241}\text{Am}$  subsequently decays to  $^{237}\text{Np}$ , also an  $\alpha$ -emitter ( $t_{1/2} = 2.1 \times 10^6$  years). In the short-term assessment of dose, either due to atmospheric testing or severe reactor accidents, the contribution of  $^{241}\text{Pu}$  is important and often underestimated. However, in the long-term management of contamination due to fall out, nuclear accidents, and transuranic (TRU) waste disposal,  $^{241}\text{Am}$  and  $^{237}\text{Np}$  present much higher risks. In this context, understanding the spatial and temporal distribution of  $^{241}\text{Am}$  in the environment, at or near reprocessing and disposal facilities, is crucial for assessing the long-term radiological

Responsible editor: Georg Steinhauser

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dose. This radionuclide is therefore of particular interest at the U.S. Department of Energy's (USDOE) WIPP, where  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  account for more than 99% of the total radioactivity slated for disposal (USDOE 2014).

The WIPP, located near Carlsbad in southeastern New Mexico, is the only operating deep geological repository for defense-related TRU and mixed transuranic waste (MTRU), i.e., waste containing both TRU and hazardous wastes, in the USA. The repository is mined into a thick Permian-age sequence of interbedded salt (halite) and anhydrite, known as the Salado Formation, ~655 m (2150 ft.) below ground surface that is essentially non-radiogenic. The facility is authorized by the WIPP Land Withdrawal Act (Public Law 102-579 n.d.) to dispose of 175,000 m<sup>3</sup> of TRU waste. Since starting operations in March 1999, some 91,000 m<sup>3</sup> of waste have been emplaced in the repository. The facility recently resumed waste disposal operations after a 3-year hiatus due to an accidental radiological release in February 2014. It is estimated that the WIPP repository will contain  $1.20 \times 10^4$  kg of Pu isotopes and 203 kg of  $^{241}\text{Am}$  at closure (USDOE 2014). The transuranic actinides are the key constituents of nuclear waste for long-term considerations as many of them are  $\alpha$ -emitters and have very long half-lives. Even though americium are less widely discussed compared to plutonium isotopes, the separation of americium from nuclear waste streams is a major goal of fuel reprocessing research. In this context, a clear understanding of the coordination and environmental behavior of the americium is essential for the safe management and disposal of radioactive waste (Welch et al. 2017; Dares et al. 2015). The stable oxidation state of Am in the environment is Am(III). However, formation of higher oxidation states of americium such as  $\text{AmO}_2$ ,  $\text{Am(V)O}_2^+$ , and  $\text{Am(VI)O}_2^{2+}$  have been reported in acidic media. Selective oxidation of trivalent americium has been used in the separation of americium from the lanthanides in nuclear waste streams (Dares et al. 2015).

Generally, as with the other transuranium elements, most of the americium present in the environment today is due to atmospheric nuclear tests that were conducted between 1945 and 1980. It is estimated that nuclear weapons testing dispersed some 13 PBq (1 PBq =  $10^{15}$  Bq) of  $^{239+240}\text{Pu}$  and 3.1 PBq of  $^{241}\text{Am}$  into the atmosphere (UNSCEAR 1982). Nuclear reactor accidents, such as Chernobyl and Fukushima, and other releases from weapons production facilities have caused localized contamination in some areas. Since  $^{241}\text{Am}$  is the  $\beta$ -decay daughter of the short-lived  $^{241}\text{Pu}$ , its concentration continues to increase in the environment. Concentrations of  $^{241}\text{Am}$  are estimated to peak 70–80 years following an accidental release of  $^{241}\text{Pu}$  (US-EPA 1976). Generally,  $^{241}\text{Am}$  and plutonium isotopes have been measured as traces in environmental samples with a  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratio of ~0.37 and  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity ratio of ~0.024 at latitudes of 40° to 50° N,

indicating their global fallout origin (UNSCEAR 2000). However, the  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratio is expected to increase with time, reaching ~0.40 by 2035 (Leon Vintro et al. 1999).

The background radiation levels at the WIPP are influenced primarily by natural radioactivity and global fallout from nuclear weapons tests with a possible contributor being anthropogenic radioactivity remaining from Project Gnome. The Project Gnome was the first nuclear test conducted in the Plowshare Program that focused on developing peaceful uses for nuclear explosives (USAEC 1973). The December 1961 test detonated a  $^{239}\text{Pu}$  device, with a 3.0-kt equivalent TNT yield, ~366 m below ground surface in a thick salt deposit. The Gnome plume reportedly traveled mostly north-northwest not too far from the WIPP site. Although it had been planned as a contained explosion, radioactive materials were vented to the surface. The Gnome site is located only 8.8 km southwest of the WIPP site and falls within the pre-operational radiological surveillance area for the WIPP. The site was decontaminated several times following the test and surface contamination is now well below levels of public health and environmental concern. However, prior to the opening of the WIPP, low levels of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ , and  $^{239+240}\text{Pu}$  were still detectable in surface soils at the Gnome site (Faller 1994; Kenney et al. 1995). These contaminated soils remain likely sources of contamination in samples of environmental media collected to demonstrate regulatory compliance.

Another likely contributor to current background radiation levels at the WIPP is a recent accidental radiological release from the repository. On February 14, 2014, a waste container in the repository underwent a chemical reaction that caused it to overheat and rupture to release radionuclides, mostly  $^{241}\text{Am}$ , into the disposal room (USDOE 2015). A small amount of radioactivity escaped to the surface through the ventilation system and was detected approximately 1 km away from the facility. Source term estimation suggests a release of  $\sim 3.7 \times 10^7$  Bq of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  into the environment, which would have briefly increased the concentration of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  in surface air in the vicinity of the WIPP site (Thakur et al. 2016).

In this context, the variation in concentrations of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  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. Since the atmospheric nuclear weapons tests of the 1950s and 1960s, plutonium, and to some extent  $^{241}\text{Am}$ , have become ubiquitous elements in the environment, including at the WIPP site. Several studies have assessed the distribution and long-term behavior of plutonium in the WIPP environment (Thakur et al. 2012; Hayes and Akbarzadeh 2014). However,  $^{241}\text{Am}$  has received much less attention than plutonium despite being the second most abundant actinide in TRU waste. In fact, there have been no studies on the temporal and spatial distribution of  $^{241}\text{Am}$  in the WIPP environment.

Radionuclides present in the environment, whether naturally occurring or anthropogenic, may result in radiation doses to humans. Therefore, environmental monitoring is required around nuclear facilities to characterize radiological baseline conditions, identify any releases, and determine the effects of releases should they occur. In this study, the variation of  $^{241}\text{Am}$  before and after the WIPP begin accepting TRU waste is assessed. Historical data, collected over more than a decade, are used to characterize the distribution and long-term behavior of  $^{241}\text{Am}$  in the WIPP environment. Isotopic ratios, derived from measurements in different environmental media, are analyzed to gain insight into likely sources contributing to the  $^{241}\text{Am}$  profile in the WIPP environment.

## Experimental

### Site description and sampling locations

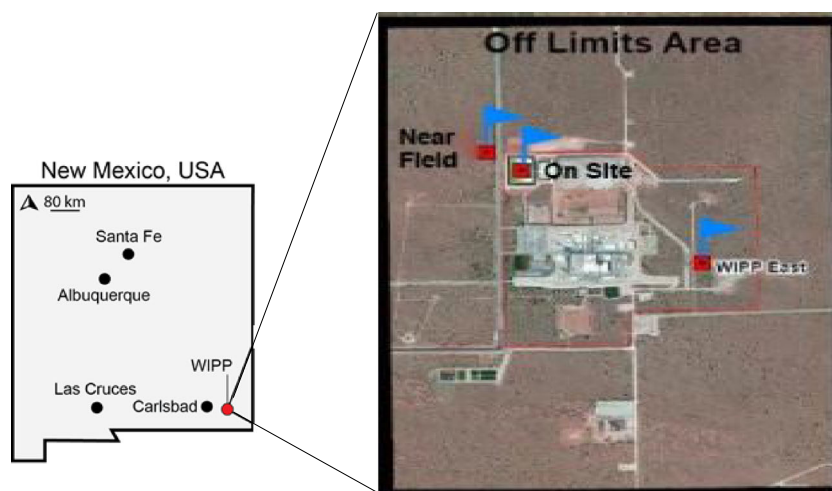
The WIPP facility, shown in Fig. 1, encompasses 41.4 km<sup>2</sup> and consists of surface buildings, an array of vertical shafts, and a mined repository ~650 m below ground surface. This part of New Mexico is relatively flat, sparsely inhabited, with little surface water. The climate in this region is classified as cold, semi-arid. Winds are predominantly from the southeast and with speeds ranging from 3.7 to 6.3 ms<sup>-1</sup>. The annual average precipitation is 324 mm and the annual average temperature is 17 °C with average highs over 40 °C in the summer months and lows around 15 °C. In the winter months, daily temperatures are cooler with highs around 10 to 15 °C and lows reaching -10 °C. The main population centers lie within 80 km of the site and are concentrated in and around the communities of Carlsbad, Hobbs, Eunice, Loving, Jal, Lovington, and Artesia, with an estimated population of 88,952.

Lands in the immediate vicinity of the WIPP and Project Gnome sites are managed by the U.S. Department of the Interior Bureau of Land Management and are used for livestock grazing, potash mining, oil and gas exploration and production, and recreational activities. Environmental monitoring, to demonstrate compliance with the regulations, includes airborne particulates, soil, surface water, groundwater, sediments, and biota.

### Airborne particulate sampling

Airborne particulate samples were collected in the vicinity of the WIPP site using high-volume air samplers at a volumetric flow rate of ~1.13 m<sup>3</sup> min<sup>-1</sup>. These samplers are installed at monitoring stations around the site (Fig. 1) and include: (1) Onsite, located about 0.1 km northwest of the WIPP exhaust shaft; (2) Near Field, about 1 km northwest of the WIPP facility; and (3) Cactus Flats, about 19 km southeast of the WIPP facility. The sampling sites were selected based on their likelihood of intercepting accidentally released airborne radionuclides using an analysis of prevailing wind direction and speed scenarios. Particulate samples were collected on pre-weighed, 20 × 25 cm A/E™ glass fiber filters with a nominal 1.0 μm pore size (Pall German Laboratory, Ann Arbor, MI). Samplers are operated to maximize particulate loading without impacting air flow. However, filters were changed when the volumetric flow rate dropped below 0.99 m<sup>3</sup> min<sup>-1</sup>, when there was a power outage, or when the sampler malfunctioned due to mechanical issues. Each filter was weighed after retrieval and the change in weight used to determine the weight of particulate material collected over the sampling interval. Actinide analyses were performed on individual filters. A typical sampling period lasts for about 3 to 4 weeks depending on the levels of particulate matter that accumulates on the filters. An average ~28,000 m<sup>3</sup> air flow through these filters.

**Fig. 1** Location of the Waste Isolation Pilot Plant (WIPP) in southeastern New Mexico, USA, including the location ambient air samplers and soil sampling areas



## Soil sampling

Soil samples were collected from the two locations (Near Field and Cactus Flats) where the high-volume air samplers are stationed (Fig. 1). Bulk soil samples were collected with a trowel from a depth of 0–2 cm. At both locations, samples were taken at random orientations and distances (0–16 km) from the stations. Individual sampling locations were selected on the basis of having relatively flat topography, minimum surface erosion, and minimum surface disturbance by human or livestock activity.

## Sample digestion

Filter particulate samples were prepared for radiochemical analyses by first ashing in a muffle furnace at 500 °C for 6 h. This was followed by wet digestion with a mixture of concentrated nitric ( $\text{HNO}_3$ ), hydrochloric ( $\text{HCl}$ ), and hydrofluoric ( $\text{HF}$ ) acids on a hot plate at 200 °C. Samples were then treated with concentrated perchloric acid ( $\text{HClO}_4$ ) and  $\text{HNO}_3$  to remove fluoride ions. The inside walls of the beaker were rinsed carefully with  $\text{HNO}_3$ , to gather residual  $\text{HF}$ , and heated on a hot plate to ensure removal of residual  $\text{HF}$  from the matrix. The residues were then dissolved in 1.0 M  $\text{HCl}$  for subsequent actinide separation and analysis. Ashing at a higher temperature may result in loss of plutonium if only the  $\text{HNO}_3$  leaching method is employed. Wang et al. (2015) observed a loss of about 40% plutonium in soil samples at ashing temperature > 450 °C. More aggressive digestion such as combination of  $\text{HNO}_3$  and  $\text{HF}$  or alkali fusion method is necessary if samples are ashed at > 500 °C. Use of glass fiber filter in this case is also warranted  $\text{HNO}_3$  +  $\text{HF}$  digestion.

Soil samples were dried at 110 °C and blended to homogenize prior to digestion. A 5-g aliquot of each homogenized sample was dissolved by heating with a mixture of  $\text{HNO}_3$ ,  $\text{HCl}$ , and  $\text{HF}$  acids. The sample residues were then heated with  $\text{HClO}_4$  and boric acid ( $\text{H}_3\text{BO}_3$ ) to remove  $\text{HF}$ . After cooling, samples were transferred to a 50-mL centrifuge tube and centrifuged at 3600 rpm for 10 min. The supernatant was filtered through a 0.45- $\mu\text{m}$  filter and transferred to a 250-mL beaker. The actinides were subsequently separated as a group by coprecipitation on  $\text{Fe}(\text{OH})_3$ .

The resins used in this work were TEVA (Aliquot 336), TRU-resin (tri n-butyl phosphate, [TBP]), N-diisobutyl carbamoyl methyl phosphine oxide (CMPO), and an anion exchange resin (Eichrom 1- $\times$ 8, 100–200 mesh, chloride form) obtained from Eichrom Technologies, Inc. Chemical recoveries were determined using  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  as yield tracers. Nitric, perchloric, and hydrochloric acids were prepared from reagent-grade acids (Fisher Scientific, Inc.). All other chemicals were ACS reagent grade and dilutions were made with de-ionized water.

## Radiochemical separation

The actinides were concentrated in an iron hydroxide precipitate as  $\text{Fe}(\text{OH})_3$ . After decantation and centrifugation, the precipitate was dissolved in 10 mL of concentrated  $\text{HNO}_3$  and diluted to 20 mL to make the solution 8 M in  $\text{HNO}_3$ . The oxidation state of plutonium as Pu(IV) was adjusted by adding 1 mL of 1 M  $\text{NH}_4\text{I}$  with a 10-min wait step, after which 2 mL of 2 M  $\text{NaNO}_2$  was added. The sample solutions were then loaded onto anion exchange columns, pre-conditioned with 8 M  $\text{HNO}_3$ . The columns were then washed with  $3 \times 10$  mL 8 M  $\text{HNO}_3$ . Finally, plutonium was eluted with 30 mL of 0.1 M  $\text{NH}_4\text{I}$  + 10 M  $\text{HCl}$ . A two-step column separation process was used to ensure complete removal of any interference.

An Eichrom-TRU column (2 mL, 100–150  $\mu\text{m}$ ), pre-conditioned with 10 mL of 2 M  $\text{HNO}_3$ , was used for the separation of americium. The fraction containing americium from the 8 M  $\text{HNO}_3$  eluate of the first anion exchange resin column was evaporated to dryness and re-dissolved in 10 mL of 2 M  $\text{HNO}_3$ . A solution of  $\text{NH}_4\text{SCN}$  was used to test for the presence of  $\text{Fe}^{3+}$ . If  $\text{Fe}^{3+}$  was present, then 300 mg of ascorbic acid was added to reduce  $\text{Fe}^{3+}$  to  $\text{Fe}^{2+}$ . The sample solutions were then loaded onto exchange columns and washed with  $3 \times 5$  mL of 2 M  $\text{HNO}_3$ . The columns were then washed with 10 mL of 2 M  $\text{HNO}_3$  + 0.05 M  $\text{NaNO}_2$ . This step was used to oxidize any  $\text{Pu}^{3+}$ , formed by the reduction of  $\text{Pu}^{4+}$  with ascorbic acid, back to  $\text{Pu}^{4+}$  to prevent any  $\text{Pu}^{3+}$  from co-eluting with the  $\text{Am}^{3+}$ . Americium was then eluted with 20 mL of 4 M  $\text{HCl}$ .

Polonium is often present in significant amounts in particulate samples. Therefore, an additional step using an anion exchange column was added to allow effective removal of polonium from the  $\text{Am}^{3+}$  fraction (Lemons et al. 2018). The americium fraction from the TRU column was evaporated to dryness with 1 mL of 50%  $\text{H}_2\text{SO}_4$ , to enhance destruction of any extractant in this solution, and then with 3 mL of  $\text{HClO}_4$ . The samples were dissolved in 4 mL of 2 M  $\text{HCl}$  and loaded onto anion exchange columns (AG1- $\times$ 8, 50–100 mesh,  $\text{Cl}^-$  form, 10 mm dia.  $\times$  5 cm long) pre-conditioned with 10 mL of 1 M  $\text{HCl}$ . The columns were washed with  $3 \times 4$  mL of 1 M  $\text{HCl}$ . The load and the washing solutions were collected in 50 mL polycarbonate centrifuge tubes.

A TEVA column was used to remove lanthanides, which are present in significant amount in soil samples, from the americium fraction. The americium fraction, stripped from the TRU columns, was re-dissolved in 10 mL of 3 M  $\text{NH}_4\text{SCN}$  + 0.1 M  $\text{HCOOH}$ , warming gently as needed. The solution was loaded onto a TEVA cartridge (2 mL, 100–150  $\mu\text{m}$ ), previously conditioned with 10 mL of 3 M  $\text{NH}_4\text{SCN}$ . The column was washed with  $3 \times 4$  mL of 1.5 M  $\text{NH}_4\text{SCN}$  + 0.1 M  $\text{HCOOH}$  to remove any lanthanides present as they interfere with alpha spectrometry peak resolution.



Americium was then eluted with 15 mL of 2 M HCl and any residual  $\text{NH}_4\text{SCN}$  destroyed by heating with 8–10 mL of  $\text{HNO}_3:\text{HCl}$  (1:3). The solution was then evaporated to dryness with 3 mL of  $\text{HClO}_4$ . The residue was dissolved in 4 mL of 2 M HCl and transferred to a 50-mL polycarbonate centrifuge tube with DI water. The individual actinides were then micro-precipitated with an Nd-carrier and HF (Hindman 1986) and counted using alpha spectrometry (Alpha Apex, Canberra).

## Results and discussion

### Temporal variations of $^{241}\text{Am}$ in the atmosphere

Americium ( $^{241}\text{Am}$ ) is not produced directly by nuclear weapons detonation, but indirectly by the decay of  $^{241}\text{Pu}$  as it disperses in the atmosphere or after terrestrial deposition. Because of high affinity of  $^{241}\text{Am}$  for particulates, the major mode of transport is atmospheric dispersion of particulate matter. Atmospheric concentrations of  $^{241}\text{Am}$  from nuclear weapons testing depend primarily on the amount of unfissioned  $^{241}\text{Am}$  and  $^{241}\text{Pu}$ . There are also contributions from lower isotopes of plutonium that are neutron activated to  $^{241}\text{Pu}$  during weapons detonation. Spatially variable concentrations arise from transport and differences in residence time, of both  $^{241}\text{Am}$  and its precursor  $^{241}\text{Pu}$ , in the various atmospheric compartments (e.g., stratosphere and troposphere). In the test of a particular weapon, this depends on the amount of plutonium in the weapon, the explosive yield, the detonation height, and meteorological conditions at the time of detonation. High-yield tests by the USA were characterized by very high ratios of  $^{241}\text{Pu}$  to  $^{239+240}\text{Pu}$ , which produced relatively large quantities of  $^{241}\text{Am}$  (Roos et al. 1994). The atmospheric residence time can span several years, long enough to allow mixing with hemispheric air to be distributed worldwide (Perkins and Thomas 1980). A typical residence time of particulate debris in the troposphere is reported to be around 30–70 days whereas it is about 12–24 months in the stratosphere (Reiter 1975). The transfer of fallout from the stratosphere to the troposphere is seasonally modulated and for the northern hemisphere, it occurs mostly in the late winter and spring with little transfer occurring during summer and autumn.

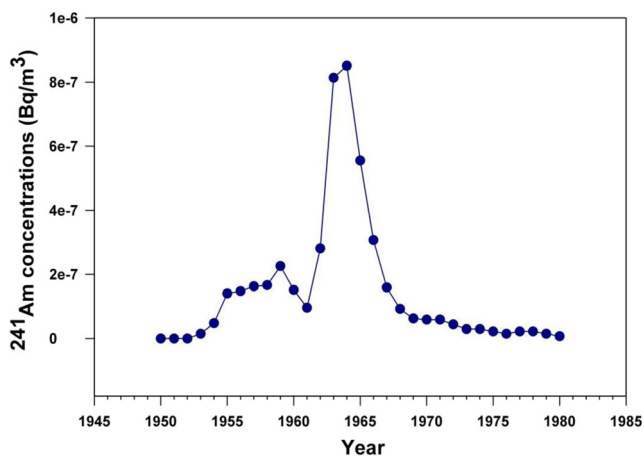
During the large-scale testing of nuclear weapons, most of the fallout debris was injected into the stratosphere. Thus, the stratosphere serves as the main reservoir for bomb-derived radionuclides in the environment. However, after the signing of the partial atmospheric test-ban treaty in 1963, the concentrations of bomb-derived radionuclides in the stratosphere decreased significantly. While the French and Chinese continued atmospheric nuclear tests until the 1980s, most of these tests were of small yields that contributed only slightly to the atmospheric radionuclide concentrations. It is generally accepted

that the current levels of plutonium and americium in the stratosphere are negligible and that most of americium in the air today is associated with resuspended soil and sediment, which were contaminated by weapons fallout.

It has been estimated that the globally deposited  $^{241}\text{Pu}$  from the nuclear weapons testing of 1950s and 1960s will ultimately produce about  $5.5 \times 10^{15}$  Bq (5.5 PBq) of  $^{241}\text{Am}$  (UNSCEAR Report 1982, Annex E). Additionally,  $\sim 1.5 \times 10^{14}$  Bq (0.15 PBq, decay-corrected to 2017) of  $^{241}\text{Am}$  has been in-grown from the decay of  $\sim 6 \times 10^{15}$  Bq ( $\sim 6$  PBq) of  $^{241}\text{Pu}$  released from the Chernobyl NPP accident in 1986 (UNSCEAR 2000, Annex J). The  $^{241}\text{Am}$  is therefore the only radionuclide whose concentration continues to increase, over time, in the environment. The activity concentrations of  $^{239+240}\text{Pu}$  in the stratosphere and troposphere (10.1–14.2 km altitude) over Switzerland, from 1973 to 1986 and from 2004 to 2011, were reported by the Alvarado et al. (2014). These authors also measured activity concentrations of  $^{241}\text{Pu}$  in high-altitude aerosol samples for the periods 1973–1977, 2007–2008, and 2010. The activity concentrations of  $^{241}\text{Am}$  during the same period in the atmosphere over Switzerland were estimated from the measured concentrations of  $^{239+240}\text{Pu}$  and the reported  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratios. There were several peaks in the stratospheric concentrations of  $^{239+240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{241}\text{Am}$  in 1970s due to the Chinese nuclear tests conducted between 1970 and 1976. After 1976, the stratospheric concentrations of these radionuclides decreased. There are no stratospheric data available for  $^{239+240}\text{Pu}$  during the period 1987 to 2003, nor for  $^{241}\text{Pu}$  between 1978 and 2006. These authors added very few new data on stratospheric plutonium isotopes ( $^{239+240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{238}\text{Pu}$ ) and  $^{137}\text{Cs}$  were added for the period between 2007 and 2010–2011. However, they found that the activity concentrations of these radionuclides in the stratosphere are about two to four orders of magnitude higher than previously assumed and suggested that the stratospheric mean residence time of these particles is 2.5–5 years.

### Temporal variations of $^{241}\text{Am}$ in surface air

There are very limited data on the surface air activity concentrations of  $^{241}\text{Am}$  in surface air. The concentrations of americium in surface air were not systematically monitored during the period 1959–1964, the time of the heaviest contributions from global fallout. The annual average concentrations of  $^{241}\text{Am}$  in surface air, prior to 1965, in the mid-latitudes of the northern hemisphere resulting from nuclear weapons testing between the 1950s and the 1980s were estimated using an atmospheric transport model and the amount of  $^{241}\text{Am}$  that would have been produced as a result of the testing (Bennett 1979). As shown in Fig. 2, surface air concentrations of  $^{241}\text{Am}$ , attributed to global fallout, increased rapidly and reached a maximum of  $0.85 \mu\text{Bq}/\text{m}^3$  in 1964. Since 1973,



**Fig. 2** Predicated surface air concentrations of <sup>241</sup>Am during 1951–1980 (From ref. Bennett 1979)

levels have been declining. It is this global fallout that created a legacy of <sup>241</sup>Am on the terrestrial surface.

### Temporal variations of <sup>241</sup>Am in surface air at the WIPP

The background concentrations of <sup>241</sup>Am in the vicinity of the WIPP site were measured by Lee et al. (1998). The aerosol samples were collected between February 23 and May 6, 1996 using high-volume samplers. Samplers concurrently obtained total suspended solids (TSP), particles > 10 μm (PM<sub>10</sub>) at a 5-m elevation, and PM<sub>10</sub> at a 2-m elevation. Activity concentrations of <sup>241</sup>Am in the samples were 0.008 ± 0.002 μBq/m<sup>3</sup> in the PM<sub>10</sub> at 2 m, 0.006 ± 0.002 μBq/m<sup>3</sup> in the PM<sub>10</sub> at 5 m, and 0.011 ± 0.001 μBq/m<sup>3</sup> in the TSP at 5 m (Lee et al. 1998). The background concentrations of <sup>239+240</sup>Pu, <sup>241</sup>Am, and other radionuclides within and adjacent to the WIPP site were also measured by the Environmental Evaluation Group (EEG) for the period between 1985 and 2000 (Gray et al. 2000). Average pre-operational baseline concentrations reported in their study were 0.027 ± 0.11 μBq/m<sup>3</sup> (n = 79) for <sup>241</sup>Am, 2.3 ± 0.056 μBq/m<sup>3</sup> (n = 88) for <sup>239+240</sup>Pu, and 0.006 ± 0.062 μBq/m<sup>3</sup> (n = 90) for <sup>238</sup>Pu. None of the composite samples were statistically different from the lower limits of detection for <sup>239+240</sup>Pu or <sup>241</sup>Am.

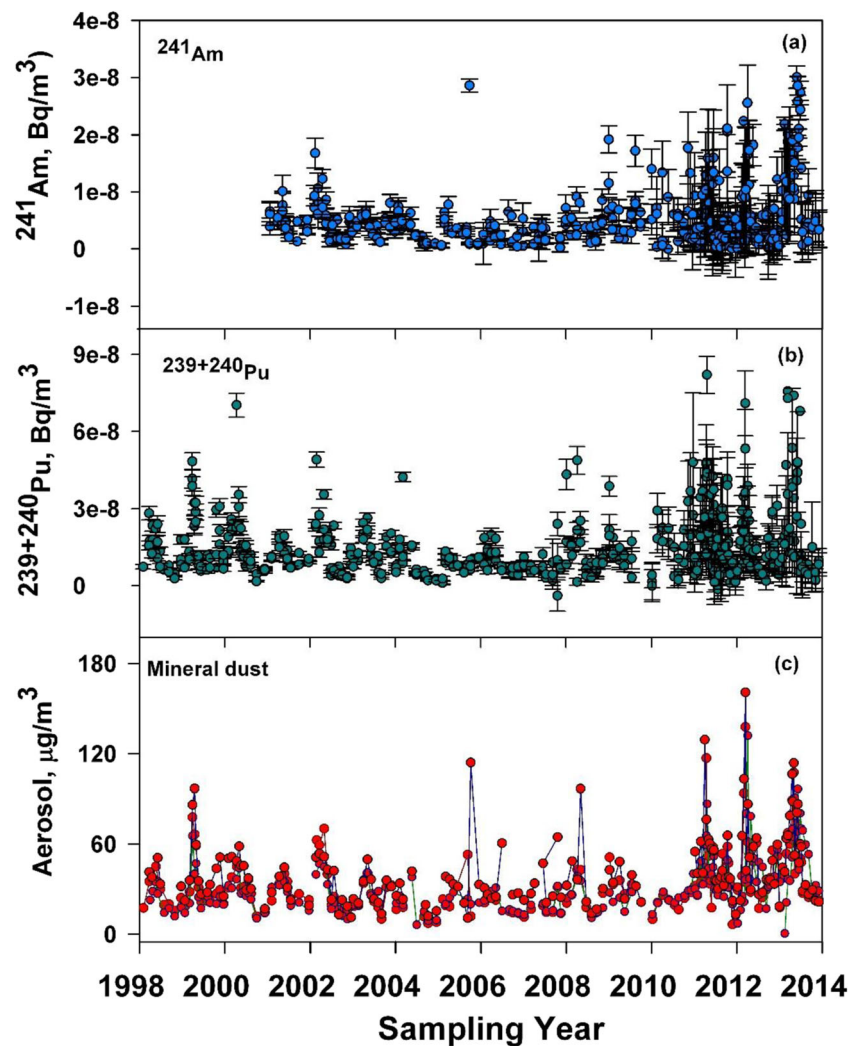
A time series of <sup>241</sup>Am airborne activity concentration, measured in aerosol filter samples collected from the Onsite, Near Field, and Cactus Flats stations from 2001 through 2013, is shown in Fig. 3a. The data show frequent detection of trace levels of <sup>241</sup>Am and <sup>239+240</sup>Pu. The activity concentrations of <sup>241</sup>Am measured in the ambient surface air, in the WIPP vicinity, range from 0.00036–0.126 μBq/m<sup>3</sup> at Onsite station, to 0.00012–0.05 μBq/m<sup>3</sup> at Near Field station, and 0.00017–0.113 μBq/m<sup>3</sup> at Cactus Flats station (CEMRC 2006). In general, <sup>241</sup>Am activities peak in the March to June timeframe, which coincides with the period of strong and gusty winds in

the area that frequently give rise to blowing dust. The activity concentrations of <sup>239+240</sup>Pu, measured in aerosol samples from the three study sites, are shown in Fig. 3b. The detection of <sup>241</sup>Am in surface air was not as frequent as that of <sup>239+240</sup>Pu because of the normally low levels of <sup>241</sup>Am in the environment. Current low levels are due to <sup>241</sup>Am resulting primarily from the decay of <sup>241</sup>Pu, rather than fallout. However, when detectable, the activity concentrations of <sup>241</sup>Am in the high-volume samples closely tracked those of <sup>239+240</sup>Pu. Most notably, strong springtime peaks in <sup>241</sup>Am activity concentrations were evident in the samples from 2001 through 2002, and from 2011 through 2013. The amount of particulate deposits (mass loadings) on filters also tends to increase during the windy period at all three stations. Figure 3b also shows that the aerosol mass loadings, i.e., the mass of aerosols collected per unit volume of air, followed a seasonal pattern similar to that of <sup>239+240</sup>Pu and <sup>241</sup>Am activity concentrations. Although seasonality in <sup>241</sup>Am data is not as pronounced as in the case of <sup>239+240</sup>Pu (Fig. 3c), the data do show a typical seasonal variation with highs in the spring and lows in the summer. The observed seasonality in <sup>241</sup>Am activity concentrations in the WIPP environment can therefore be attributed to the resuspension of contaminated soil and sediment.

Temporal variations in surface air concentrations of <sup>239+240</sup>Pu and <sup>241</sup>Am activity concentrations have been observed since the early days of atmospheric monitoring. This cycle was confirmed by numerous data collected in the northern hemisphere. For example, Arnold and Wershofen (2000) showed seasonality in <sup>239+240</sup>Pu concentrations in ground-level air from Germany. Salminen and Paatero (2009) observed a seasonal cycle in <sup>239+240</sup>Pu, <sup>241</sup>Pu, <sup>241</sup>Am, and <sup>238</sup>Pu concentrations in surface air of Sodankyla, Finland in 1963. A springtime enhanced activity of <sup>239+240</sup>Pu was reported in the dry and wet deposition samples collected from Tsukuba, Japan, and Daejeon, South Korea (Hirose et al. 2003; Hirose et al. 2004). While the current americium/plutonium aerosol data (post-1984) and those collected during the era of atmospheric nuclear weapons testing (pre-1984) both show springtime peaks, the causes for the cycles are likely quite different. It has been suggested that the seasonal cycle in aerosol samples collected prior to the cessation of nuclear weapons testing is associated with a stratospheric-tropospheric exchange phenomenon, while the resuspension is believed to be the main source of <sup>241</sup>Am in aerosol samples collected after the cessation of nuclear weapons testing.

In general, the <sup>241</sup>Am activity concentrations in the ambient surface air after WIPP became operational are not statistically different from those measured prior to waste disposal operations. The exception is 2014 when higher-than-normal background levels of <sup>241</sup>Am and <sup>239+240</sup>Pu were detected in February and March, 2014. The increased activity concentrations are attributed to an accidental release from the repository

**Fig. 3** Temporal patterns in atmospheric activity concentrations of  $^{241}\text{Am}$  (a),  $^{239} + ^{240}\text{Pu}$  (b), and aerosols mass loading on the filters (c) at three stations in the vicinity of the WIPP site



due to a drum breach on February 14, 2014 (USDOE 2015). This event released moderate levels of radioactivity, mostly  $^{241}\text{Am}$  and  $^{239} + ^{240}\text{Pu}$ , into the underground air. A small amount of radioactivity escaped to the atmosphere through the ventilation system and was detected at the surface. The highest activity concentrations detected were  $115.2 \mu\text{Bq}/\text{m}^3$  for  $^{241}\text{Am}$ , and  $10.2 \mu\text{Bq}/\text{m}^3$  for  $^{239} + ^{240}\text{Pu}$  at Onsite station, which is only 0.1 km northeast of the exhaust shaft. At Near Field station, which is 1 km northwest of the exhaust shaft (i.e., the predominant wind flow direction), the activity concentrations were  $81.4 \mu\text{Bq}/\text{m}^3$  for  $^{241}\text{Am}$  and  $5.78 \mu\text{Bq}/\text{m}^3$  for  $^{239} + ^{240}\text{Pu}$ . A third station at Cactus Flats, located 19 km southeast (upwind) of the shaft, showed no increase in  $^{241}\text{Am}$  or  $^{239} + ^{240}\text{Pu}$  (CEMRC 2014). While levels of  $^{241}\text{Am}$  and  $^{239} + ^{240}\text{Pu}$  were above the pre-release background levels, it is important to note that these concentrations were below any level of public health or environmental concern. It should also be noted that the sampling rate for high-volume samplers,  $1.13 \text{ m}^3$  (40 cfm), is significantly greater than the  $0.03\text{--}0.14 \text{ m}^3$  (1–5 cfm) respiration rate for humans so any attempt

to estimate internal dose from exposure to the reported levels would need to account for the volume differences. The regulatory limit for exposure to a standard adult member of the public from activities on the WIPP site is 1 mSv/year based on 10 CFR 835. According to DOE, the maximally exposed general member of the public would have received a dose of less than 0.34 mSv/year (34 mrem/year) in 2014 from the WIPP operations (USDOE 2014). The average person living in the USA receives an annual dose of about 6.2 mSv (620 mrem) from exposure to naturally occurring and medical sources of radiation (Thurston 2010).

Americium data from the WIPP environment were also compared with other national and international monitoring data to assess the worldwide concentrations of  $^{241}\text{Am}$  in surface air. At the Hanford Site in eastern Washington, USA, background concentrations of  $^{241}\text{Am}$  measured ranged from  $0.015$  to  $0.033 \mu\text{Bq}/\text{m}^3$  during the 1993–1994 period. These are significantly lower than the range of  $0.20\text{--}1.1 \mu\text{Bq}/\text{m}^3$  in the 100-Hanford's K Area, and  $0.12\text{--}0.93 \mu\text{Bq}/\text{m}^3$  in the 200-East Area of the Hanford site measured during the 1999–2001

period (Poston et al. 2002). At the Los Alamos National Laboratory site in northern New Mexico, USA, the highest concentration measured was  $0.25 \mu\text{Bq}/\text{m}^3$  (quarterly composite) over the period 1986–1997 (Eberhart 1998). In Vilnius, Lithuania, measured  $^{241}\text{Am}$  concentrations ranged from  $0.0003$  to  $0.5 \mu\text{Bq}/\text{m}^3$  during the 1995–2003 periods compared to a range of  $0.0005$  to  $0.025 \mu\text{Bq}/\text{m}^3$  during the 2005–2006 periods (Lujaniené et al. 2012a). Surface air samples collected in 1963 from Sodankylä, Finland and measured in 2007 for the in-growth of  $^{241}\text{Am}$  from the  $^{241}\text{Pu}$  initially collected on these filters had  $^{241}\text{Am}$  concentrations ranging from  $<0.5$  to  $50 \mu\text{Bq}/\text{m}^3$  (Salminen and Paatero 2009). Lehto et al. (2006) reported  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  concentrations of  $1400 \mu\text{Bq}/\text{m}^3$  and  $0.014$ – $2.5 \mu\text{Bq}/\text{m}^3$ , respectively, in surface air at Kurchatov, Kazakhstan (near ground zero) during the 2000–2001 timeframe.

A brief increase in surface air concentrations of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  was reported by many monitoring stations around Europe during 1986–1987 due to the Chernobyl accident. For example, average surface air concentrations of  $^{241}\text{Am}$  in Roskilde, Denmark were in the range  $5.2$ – $11.0 \mu\text{Bq}/\text{m}^3$  during April–May, 1986 (Aarkrog 1988a). Levels measured in Austria were in the range  $7.4$ – $10.4 \mu\text{Bq}/\text{m}^3$  (Irlweck and Wicke 1998). In Bragin, located about 55 km north of Chernobyl, the mean activity concentration of  $^{241}\text{Am}$  during this period was  $40 \mu\text{Bq}/\text{m}^3$  (Knatko et al. 1993). Similarly, elevated levels of  $^{239+240}\text{Pu}$  in the range  $10$ – $28 \mu\text{Bq}/\text{m}^3$  were measured in Prague (Holgye and Filgas 1987);  $1.2$ – $89 \mu\text{Bq}/\text{m}^3$  in Vienna (Holgye and Filgas 1987);  $32 \mu\text{Bq}/\text{m}^3$  in Nurmijärvi, Finland (Paatero et al. 2010); and  $0.4$ – $10.6 \mu\text{Bq}/\text{m}^3$  in Belgrade, Serbia (Manić-Kudra et al. 1995). However, following a peak in 1986, the concentrations of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  diminished. The activity concentrations of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  measured in Vilnius, Lithuania in March–April, 2011 were  $0.016 \mu\text{Bq}/\text{m}^3$  and  $0.044 \mu\text{Bq}/\text{m}^3$ , respectively (Lujaniené et al. 2012b).

### Temporal variability of $^{241}\text{Am}$ activity due to resuspension in the vicinity of WIPP

Given the length of time since the cessation of nuclear weapons testing, most of the  $^{241}\text{Pu}$  deposited on the land surface near the WIPP is expected to be fully decayed into  $^{241}\text{Am}$  and incorporated into the soil. This is the basis for the hypothesis that americium detected in aerosol samples is associated with resuspended soil particles. Resuspension is recognized as the predominant mechanism for maintaining residual plutonium and americium in surface air. Most radioactive material transported by wind is a result of saltation, which occurs at a typical threshold wind speed ranging from  $6$  to  $13 \text{ms}^{-1}$  at  $0.3 \text{m}$  (Stout and Arimoto 2010; Arimoto et al. 2005). Under these conditions, particles less than  $50 \mu\text{m}$  may be suspended for extended periods of time. The effectiveness of wind

transport in spreading radionuclides across the landscape also varies with particle size. For example, at the Hanford site,  $^{241}\text{Am}$  was reported to have been transported in the air on different particle sizes and reached maximum concentrations at different heights than those of  $^{239+240}\text{Pu}$  (Sehmel 1987). Over time, radionuclide concentrations decrease rapidly, especially at high wind speeds (Hollander 1994). Arimoto et al. (2005) have shown that under semi-arid conditions like those at the WIPP, plutonium-bearing aerosols tend to increase in ambient air samples as wind speeds approach  $4 \text{ms}^{-1}$ , reaching a maximum at  $7 \text{ms}^{-1}$ , and staying constant at wind speeds above  $7 \text{ms}^{-1}$ . These aerosols are readily trapped on the filter in an air monitoring station, leading to their frequent radionuclide detection during high-wind events.

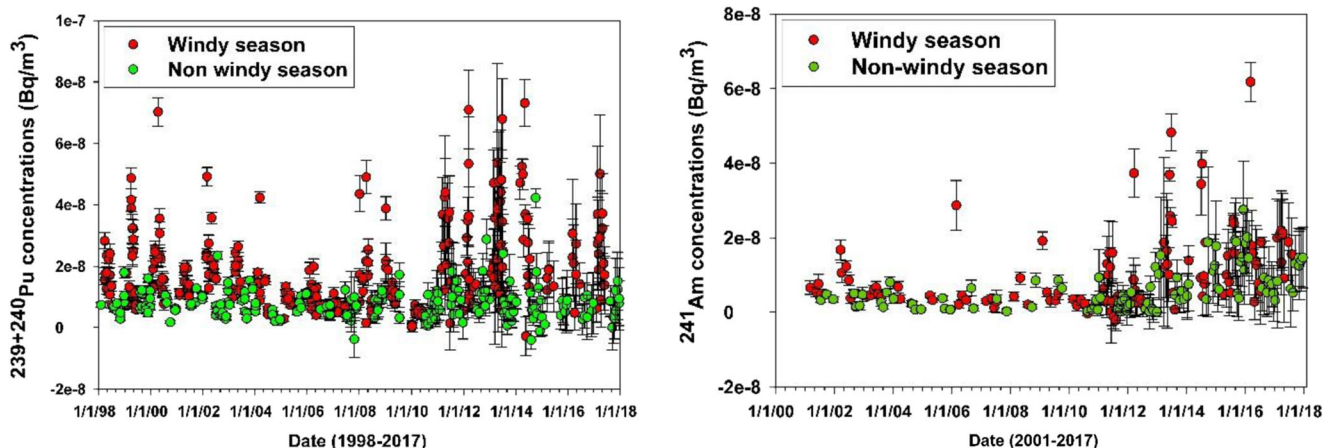
Positive detections of  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  in aerosol samples, from the three study sites, during the March to June timeframe when wind gusts are more common, are attributed to resuspension. As shown in Fig. 4, about 60–70% of positive  $^{239+240}\text{Pu}$  or  $^{241}\text{Am}$  detections are sampled during the gusty period. The apparent seasonality in plutonium activity concentrations is therefore due to the resuspension of contaminated soil particles. A linear relationship between the mass of dust retained on the filters and the  $^{239+240}\text{Pu}$  ( $r^2 = 0.68$ ) and  $^{241}\text{Am}$  ( $r^2 = 0.69$ ) concentrations is also evident at the three monitoring stations (Fig. 5).

These findings suggest that the detection of  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  in the aerosol samples collected from ambient air around the WIPP facility is primarily due to the resuspension of contaminated soils. In the WIPP environment, the concentrations of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  were similar at all stations and a good correlation exists between  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  concentrations ( $r^2 = 0.70$ ) even though neither  $^{239}\text{Pu}$  nor  $^{240}\text{Pu}$  are the immediate progeny of  $^{241}\text{Am}$  (Fig. 6).

### Near-surface distribution of $^{241}\text{Am}$ in the WIPP soils

Surface soil concentrations of radionuclides are the primary source of nearby and downwind aerosol concentrations. Therefore, it is important to understand near-surface soil concentrations of  $^{241}\text{Am}$  at and around the WIPP site. Based on data collected after the WIPP become operational,  $^{241}\text{Am}$  concentrations in the Near Field soil ranged from  $0.002$  to  $0.14 \text{Bq}/\text{kg}$ , with a mean value of  $0.044 \text{Bq}/\text{kg}$ . The same dataset showed  $^{241}\text{Am}$  concentrations in a range from  $0.007$  to  $0.26 \text{Bq}/\text{kg}$ , with a mean value of  $0.068 \text{Bq}/\text{kg}$  in the Cactus Flats soil. The corresponding concentrations of  $^{239+240}\text{Pu}$  at these two locations ranged from  $0.001$  to  $0.40 \text{Bq}/\text{kg}$ , with a mean of  $0.11 \text{Bq}/\text{kg}$  in the Near Field soil, and  $0.013$  to  $0.51 \text{Bq}/\text{kg}$ , with a mean of  $0.2 \text{Bq}/\text{kg}$  in the Cactus Flats soil. Background concentrations have been established based on soil surveys conducted at these two locations during the 1996–1997 timeframe, prior to arrival of TRU wastes at the WIPP. Background concentrations of  $^{241}\text{Am}$  are in the range





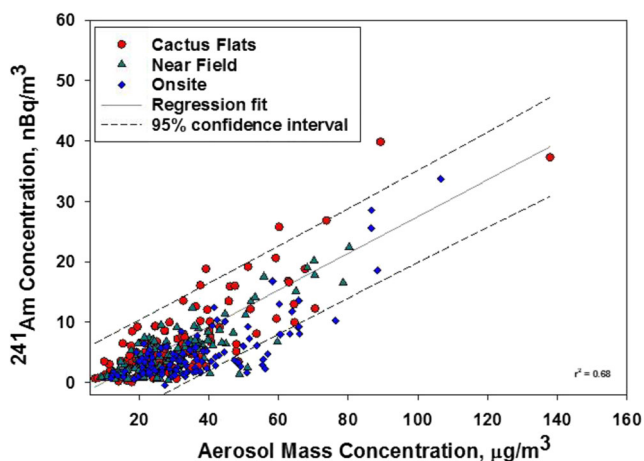
**Fig. 4** Concentrations of  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  in aerosol samples from ambient air collected during windy and non-windy seasons in the vicinity of the WIPP

0.075–0.11 Bq/kg ( $n = 9$ ), whereas background  $^{239+240}\text{Pu}$  concentrations are in the range 0.037–0.30 Bq/kg ( $n = 16$ ). A good correlation ( $r^2 = 0.64$ ) between  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  concentrations at the two locations suggests an identical origin (Fig. 7).

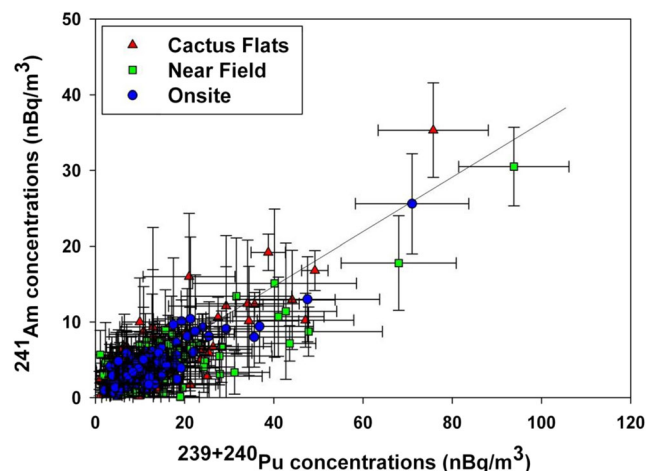
Although the concentration of  $^{241}\text{Am}$  in the surface soil at Cactus Flats is slightly higher than that in the surface soil at Near Field, there is no statistical difference between the  $^{241}\text{Am}$  concentrations at these two locations. Furthermore, there is no difference between the concentrations in soil samples collected before and after WIPP started receiving TRU waste. The deposition of radioactive fallout from weapons testing is also known to vary with latitude, being highest in middle latitudes of the northern hemisphere (UNSCEAR 1982). The range of  $^{241}\text{Am}$  concentrations in the Near Field soil is lower than background concentrations (0.6–1.7 Bq/kg) found in the Colorado Front Range, between Fort Collins and Colorado Springs (Hodge et al. 1996). The  $^{241}\text{Am}$  concentrations in the WIPP vicinity are also lower than the background range

of 0.037–1.14 Bq/kg in soils at Rocky Flats, Colorado (Hulse et al. 1999) and the 0.74–107.3 Bq/kg range in soils around the Los Alamos National Laboratory in northern New Mexico. The  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  levels in soil at various locations are summarized in Table 1.

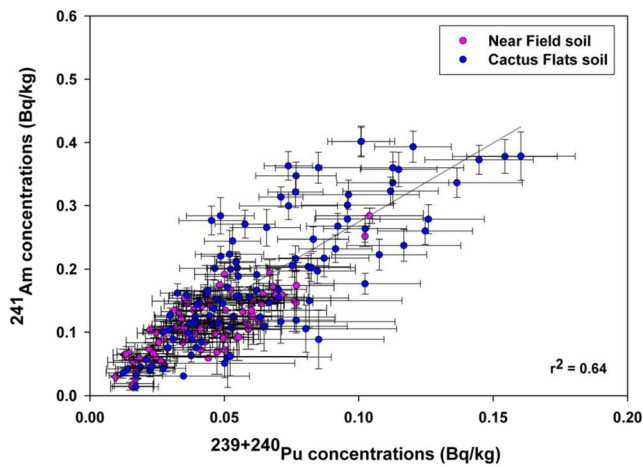
Several studies have been conducted to assess the residual contamination levels at the Project Gnome site. The EEG conducted a soil survey of the area around the Gnome site over a 7-month period in 1994–1995 (Kenney et al. 1995). In 2006, CEMRC conducted a separate study in an attempt to determine concentrations and precise isotopic ratios of Gnome contamination. Both the EEG soil survey conducted 1994–1995 (Kenney et al. 1995) and the later study by CEMRC (CEMRC 2006) reported heterogeneous distributions of  $^{241}\text{Am}$  and plutonium isotopes in the near-surface soil. Concentrations of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  reported by EEG ranged from 0.59 to 7600 Bq/kg and 4.4 to 48,000 Bq/kg, respectively. The survey by CEMRC, some 6 years later, showed concentrations ranging from 0.043 to 346 Bq/kg for  $^{241}\text{Am}$  and 0.073 to 1550 Bq/kg



**Fig. 5** A cross plot of  $^{241}\text{Am}$  concentrations and mass loading on the filters



**Fig. 6** A cross plot of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  in aerosol samples collected in the vicinity of the WIPP site



**Fig. 7** A cross plot of <sup>241</sup>Am and <sup>239+240</sup>Pu in soil samples collected in the vicinity of the WIPP site

kg for <sup>239+240</sup>Pu. The large variation in radionuclides concentration is attributed to a spotty and inconsistent clean-up effort at the site (Faller 1994).

### Depth distribution of <sup>241</sup>Am in WIPP soils

Radionuclides deposited at the soil surface in the beginning are eventually transported downward into the soil profile. The depth distribution of <sup>241</sup>Am and <sup>239+240</sup>Pu was determined, using soil samples collected at and near the WIPP site, to evaluate vertical pattern of these radionuclides in a semi-arid environment. The depth distribution of <sup>137</sup>Cs was also investigated because concentrations of <sup>137</sup>Cs in soils, from atmospheric nuclear weapons testing, are considerably greater than <sup>241</sup>Am and <sup>239+240</sup>Pu concentrations. In addition, <sup>137</sup>Cs being a gamma-emitter requires less effort for analysis. Although chemically quite different, <sup>137</sup>Cs is expected to behave similarly to <sup>239+240</sup>Pu in soils because it adheres tightly to cation-exchange surfaces and is relatively inert chemically (Coppinger et al. 1991) making it a good surrogate when the radionuclides of interest are also present.

As shown in Fig. 8, the highest concentrations of <sup>241</sup>Am and <sup>239+240</sup>Pu in soil profiles occurred at the 2 and 3 cm depths, respectively. The concentrations of these radionuclides drop off sharply within 10 cm of the soil surface and

**Table 1** The <sup>241</sup>Am and <sup>239+240</sup>Pu concentrations (Bq/kg) in soil at various locations

Background	Year	Soil depth (cm)	<sup>241</sup> Am conc. (Bq/kg)	<sup>239+240</sup> Pu conc. (Bq/kg)	References
WIPP background soil	1996–1997	0–2	0.075–0.11	0.037–0.30	CEMRC, 1998
Austrian soil					Srncik et al. 2008
Soil around REZ, NRC	1999–2000	0–20	0.056–0.94	0.064–2.0	Holgye et al. 2004
Bulgaria soil, Northern	2006	0–5	0.021–0.30	0.028–0.93	Popov et al. 2010
Bulgaria soil, Southern	2006	0–5	0.013–0.422	0.019–1.67	Popov et al. 2010
Rocky Flats, Colorado Front Range	1993–1994	0–3	0.8–1.7	–	Hulse et al. 1999
Rice field, Japan	1963	12–20	0.03–0.34	0.08–1.03	Yamamoto et al. 1983
Urbino (Marche region) Central Italy, uncultivated soil	1997	0–5	0.05–0.57	0.21–1.53	Guogang et al. 1998
Urbino (Marche region) Central Italy, cultivated soil	1997	0–5	0.012–0.086	0.046–0.22	Guogang et al. 1998
Hanford, Offsite	1971–1984	0–3	–	0.012–1.37	Posten et al. 2002
Romanian soil	1996	0–3	0.7–1.5	1.5–9.4	Breban et al. 2003
Rocky Flats, background soil	1969–1970	0–3	0.037–1.14	0.43–2.3	Litaor and Allen 1996 Poet and Martell 1972
Colorado Front Range	1995	0–5	0.4–0.9	–	USDOE 1995
Tatra mountain soil, Poland	2002–2003	0–5	0.2–3.44	0.11–17.4	Mietelski et al. 2007
SW Svalbard	2007	0–20	0.04–10.7	0.05–2.15	Łokas et al. 2013
Contaminated sites					
Mururoa Atoll	1976	12–20	0.006–0.058	0.17–0.97	
Faucon, Mururoa Atoll	1997	0–16	1.71–102.7	95–4814	Irlweck and Hmcecek 1999
Fangataufa Atoll	1997	Top soil	1.8–3.50	130–292	Irlweck and Hmcecek 1999
Palomares, Spain	1997	0–11	0.16–6.39	0.60–92.7	Irlweck and Hmcecek 1999
Rocky Flats	1997	0–3	9.2–17,100	30.2–112,900	Irlweck and Hmcecek 1999
Rocky Flats	–	0–2	0.037–10,004	1.85–53,650	Litaor and Allen 1996 Litaor 1995
Hanford Onsite	1971–1984	0–3	–	0.23–31	Posten et al. 2002
BOMARC Missile Site		0–2	0.09–1.08	0.42–6.34	Lee and Clark 2005
Semipalatinsk-21 Ground Zero		0–5	520	27,900	Yamamoto et al. 1996
Semipalatinsk-21, 1.1 km from ground zero		0–5	77	1359	Beasley et al. 1998
Maralinga, South Australia		0–5	4.0–4400	–	Cooper et al. 1994

are at or below minimum detectable concentrations (MDC) by 20 cm. There is no indication that radionuclides have been transported or redistributed to any substantial degree within these profiles. These show ~86% of  $^{137}\text{Cs}$ , 90% of  $^{239+240}\text{Pu}$ , and 78% of  $^{241}\text{Am}$  are retained in the top 5 cm of soil around the WIPP site (CEMRC 2006). These results, coupled with soil column experiments, suggest that leaching and colloidal transport are not major transport mechanisms in the redistribution of surface-deposited radionuclides in WIPP soils. There has been no analysis of the depth distributions of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  in soils at the Project Gnome site.

There have been several studies of the vertical distribution of actinides in soil profiles contaminated as a result of nuclear fallout and/or accidental releases. Generally, these studies show higher concentrations in surface soils than in subsurface soils. Several authors have also reported exponentially decreasing relationships between plutonium and americium concentrations and soil depth (Turner et al. 2003; Łokas et al. 2013). Evidence of strong retardation can be seen with  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  in Rocky Flats soils and in soil profiles collected from an undisturbed area near the Nevada Test Site (Turner et al. 2003; Hulse et al. 1999). Retardation in the near-surface is also evident in upwind and downwind soils in the Hanford Site study of Price (1991). The author reported that 98–100% of the  $^{137}\text{Cs}$ , and 95–97% of the  $^{239+240}\text{Pu}$  remained in the top 5 cm of the soil profiles. The depth profile study of  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  in sandy loam soil from the northeastern USA, conducted in 1972, also shows decreasing activity concentrations of these radionuclides with increasing soil depth (Bennett 1979). Although most of the  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  was found in the top 5 cm, measureable amounts had reached the 20-cm depth with trace amounts observed in the 20- to 30-cm depth (Bennett 1979). Subsequent measurements at the same location in 1976 show a redistribution of activity to greater depths below which it levelled off. Soil profile studies of  $^{241}\text{Am}$ ,  $^{238}\text{Pu}$ , and  $^{239+240}\text{Pu}$  at the Mururoa and Fagataufa Atolls, where French nuclear weapons tests were conducted over the period 1977

to 1974, show that more than 95% of the plutonium and americium remained in the top 5-cm layer (Irlweck and Hrnccek 1999). The profile of  $^{241}\text{Am}$  at Taranaki, a site at Maralinga in Southern Australia used for UK atomic weapons tests, also showed the majority of americium (77–96%) remained in the top 0–1 cm of soil in undisturbed areas (Burns et al. 1995; Cooper et al. 1994). In the few samples in which this is not the case, the top 1–2 cm is found to contain bulk of the activity.

High organic matter content in the near-surface is known to retain  $^{241}\text{Am}$  in the top soil layers. Boulyga et al. (2003) found that 80–95% of the plutonium and americium remained in the top 5-cm layer in turf podzol soil collected from Belarus (8–45 km to the north and northwest of Chernobyl NPP). In forest soils, in which organic matter content is typically high, ~76% of  $^{241}\text{Am}$  was found in the 0–5 cm layer (Mietelski et al. 2007). The retention of  $^{241}\text{Am}$  in the surface layer of organic-rich soil is also evident in data from the French Mercantour wetlands (Solovitch-Vella et al. 2007). Significant levels of  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  (5–10 kBq/kg) have also been found in surface gley soils of West Cumbria in Northwest England, with the actinides enriched in the organic fraction (Livens and Singleton 1991).

Some studies have reported differences in the mobilities of  $^{241}\text{Am}$  and of  $^{239+240}\text{Pu}$  in soil. For example, in an acidic, sandy soil with low clay content from the Vosges Mountains in Eastern France,  $^{241}\text{Am}$  migrated at a rate one order of magnitude higher than that of  $^{239+240}\text{Pu}$  (Solovitch-Vella, 2007). Analysis of the vertical migration of  $^{241}\text{Am}$  and plutonium in Kapachi soils, near Chernobyl and in Eastern Europe, has shown that more than 80% of Chernobyl-derived  $^{241}\text{Am}$  and plutonium remains in the top 2 cm.  $^{241}\text{Am}$  appears to migrate faster than plutonium isotopes (Mboulou et al. 1998). These data suggest that while radionuclides migrate vertically through soils, transport generally does not extend much deeper than 20 cm unless the soils are disturbed (Anspaugh et al. 1975). Depending on soil properties, recharge rates, and pore water chemistry, surface-deposited radionuclides can take years to migrate just a few centimeters.

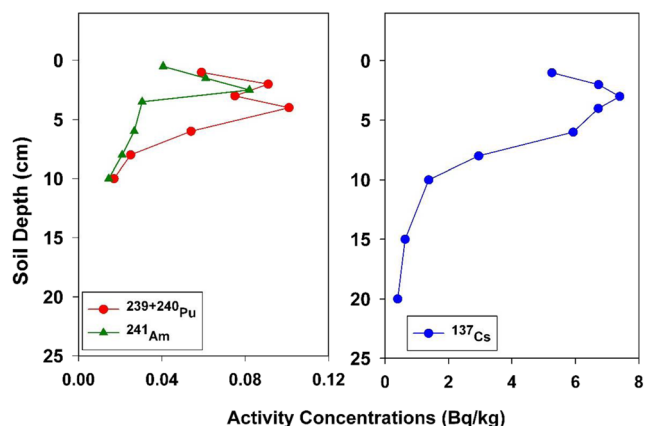


Fig. 8 Depth profiles of  $^{241}\text{Am}$ ,  $^{239+240}\text{Pu}$ , and  $^{137}\text{Cs}$  in WIPP soils

### Radionuclide activity and atom ratios in the WIPP vicinity

The  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratios measured on samples collected at the three aerosol stations, for more than a 10-year time span, are shown in Fig. 9. As of December 2013, the mean activity ratios of  $^{241}\text{Am}/^{239+240}\text{Pu}$  at the three aerosol stations were  $0.38 \pm 0.05$  (range 0.30–0.42) at Onsite station,  $0.37 \pm 0.03$  (range 0.33–0.44) at Near Field station, and  $0.39 \pm 0.03$  (range 0.35–0.44) at the Cactus Flats station. In soil samples, the mean activity ratio of  $^{241}\text{Am}/^{239+240}\text{Pu}$  was quite comparable to those measured in aerosols. In the Near Field soil, the mean  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratio was 0.42

$\pm 0.05$  (range 0.34–0.49) whereas it was  $0.39 \pm 0.07$  (range 0.29–0.51) in the Cactus Flats soil. The slightly higher range and mean in the aerosol samples are not surprising given the higher concentration of high-surface area materials that would be transported by wind compared to near-surface soil samples. Even with the differences in particle distributions of the soils and aerosols, the similarity in  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratios in the two studies is indicative of a connection between contaminated soil and aerosol. At the Gnome site, 8.8 km southwest of the WIPP, soil samples showed a  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratio of  $0.26 \pm 0.04$  ( $n = 3$ ; range 0.23–0.30), as reported by CEMRC (2006), and  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratio of  $0.22 \pm 0.12$  ( $n = 9$ ; range 0.12–0.50), as reported by Keeny et al. (1995).

The ratios in the WIPP samples are in agreement with the mean activity ratio of these radionuclides in soils and sediments reported in different studies, indicating their global fallout origin (Table 2). For example, Jia et al. (1999) measured mean ratios of  $0.35 \pm 0.05$  (range 0.23–0.43) in cultivated soil and  $0.32 \pm 0.06$  (range 0.21–0.42) in uncultivated soil collected in 1997 from Urbino in the Marche Region of Central Italy. Bunzl and Kracke (1988) measured a ratio of 0.30 in soil from Southern Germany. Data from Prague, Czech Republic, showed average ratios of 0.52 for a forest soil, 0.47 for an uncultivated soil, and 0.48 for cultivated soil near the nuclear research center at Řež (Holgye et al. 2004). Breban et al. (2003) reported ratios of 0.43–0.87 from data collected at the Carpathian Mountains, Romania, in 1996. Measurements in South Korean soils during 1992–1994 showed a ratio of 0.43 (Sha et al. 1991). Leon Vintro et al. (1999) reported that in mid-latitude soils, the  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratio was 0.37 in the late 1980s and estimated that it would reach  $\sim 0.40$  in 2000s, which is consistent with observations in the WIPP aerosol studies. Taking into account the contribution of  $^{241}\text{Am}$  from  $^{241}\text{Pu}$  decay, the observed  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratio confirms a global fallout origin.

Perturbations in the  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratios were observed in the aerosol samples of ambient air collected following the

2014 accidental radiation release at the WIPP. Both the Onsite and Near Field aerosol sampling stations detected elevated levels of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  as well as elevated  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratios. Early post-event analysis of WIPP environmental samples showed that the  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratio varied between 2.95 and 20.67 as the event progressed, but the average ratios remained fairly constant at about 10. Over the time period 2014–2016, the  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratio at Onsite station ranged from 3.06 to 11.31, whereas it ranged from 1.39 to 15.44 at Near Field station. These high activity ratios indicate the presence of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  particles associated with the breached waste drum that caused the release, and possibly the associated clean-up activities that followed.

The  $^{238}\text{Pu}/^{239+240}\text{Pu}$  ratios measured in aerosols, at and near the WIPP site, varied from 0.069 to 0.18 with a mean value of  $0.12 \pm 0.04$ . The ratio in WIPP soil ranged from 0.06 to 0.19 with a mean value of  $0.14 \pm 0.05$ . Mean ratios of  $^{238}\text{Pu}/^{239+240}\text{Pu}$  measured on soil samples from the Project Gnome site varied from  $0.14 \pm 0.025$  ( $n = 8$ ; range 0.06–0.28), as reported by CEMRC (2006) and  $0.16 \pm 0.02$  ( $n = 9$ , range 0.14–0.18), as reported by Keeny et al. (1995). These values are clearly higher than the global fallout ratio of  $\sim 0.024$  (Table 2). There is no logical explanation for these high  $^{238}\text{Pu}/^{239+240}\text{Pu}$  ratios. However, they could be attributed to the sporadic detection of  $^{238}\text{Pu}$  because of the much lower levels of  $^{238}\text{Pu}$  normally found in the environment compared to  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$ . Only around 35% of the  $^{238}\text{Pu}$  present in the environment originated from weapons testing fallout with the remaining 65% coming from that released by the burn-up of the nuclear-powered satellite SNAP-9A in April 1964 over the South Pacific (Hardy et al. 1973). Studies have shown that the SNAP-derived  $^{238}\text{Pu}$  activity in surface air peaked 2–3 years later, depending upon the latitude, and was largely depleted from the atmosphere by 1971 (Perkins and Thomas 1980).

The  $^{240}\text{Pu}/^{239}\text{Pu}$  isotopic ratio of Project Gnome soils is distinctly different from that of WIPP soils. Gnome-contaminated soils had a  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio of  $0.114 \pm 0.013$  (range 0.07–0.168) (CEMRC 2006). Soils sampled from around the WIPP site showed a mean ratio of  $0.175 \pm 0.005$  (range 0.193–0.146), whereas the global fallout ratio, based on a worldwide survey of terrestrial soils in 1970–1971, between 30 and 60° N, ranges from 0.12 to 0.21 with a mean of  $0.176 \pm 0.014$  (Krey et al. 1976). The one exception in the northern hemisphere includes soils in the southwestern USA where fallout from the Nevada Test Site was also deposited. The  $^{240}\text{Pu}/^{239}\text{Pu}$  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 tend to have higher  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios than the fallout produced by low-yield tests. For example,  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios ranging from 0.21 to 0.36 have

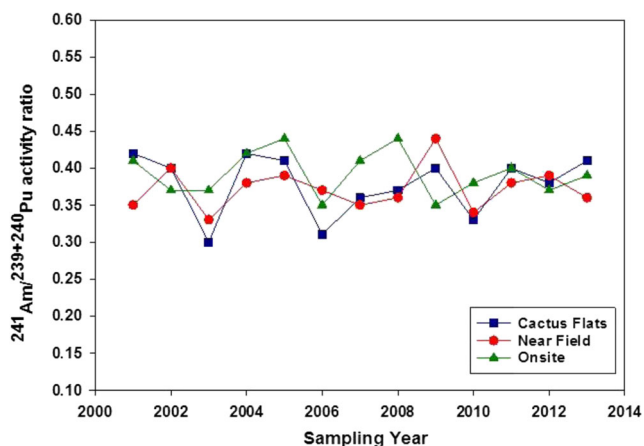


Fig. 9 Temporal variability in the  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratio measured in aerosol samples collected in the vicinity of the WIPP site



**Table 2**  $^{241}\text{Am}/^{239+240}\text{Pu}$  and  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity ratios in environmental samples

Location	Media	Sampling year	$^{241}\text{Am}/^{239+240}\text{Pu}$	$^{238}\text{Pu}/^{239+240}\text{Pu}$	References
Global Fallout, Northern hemisphere +SNAP 9A	Soil/air	1973	0.25–0.40	0.026–0.029	Baskaran et al., 1995 Hardy et al. 1973 Perkins and Thomas 1980
WIPP Site	Aerosol	1998–2013	0.37–0.39	0.12 ± 0.04 (0.069–0.18)	Present work
WIPP Site	Soil	1998–2016	0.39–0.42	0.14 ± 0.05 (0.06–0.19)	Present work/CEMRC Report 2006
Gnome Site, New Mexico	Soil	2006	0.30 ± 0.07 (0.23–0.42)	0.14 ± 0.025 (0.06–0.28)	CEMRC Report 2006
Northern Bulgaria	Soil	2006	0.415 ± 0.092 (0.30–0.60)	0.061 ± 0.022 (0.033–0.108)	Popov et al. 2010
Southern Bulgaria	Soil	2006	0.386 ± 0.073 (0.25–0.50)	0.046 ± 0.024 (0.026–0.110)	Popov and Mihailova 2010
Romania	Soil	1996	0.31–0.87	0.05–0.427	Breban et al. 2003
SW Svalbard	Peat	2007	0.46 0.10–1.5	0.023 (0.02–0.09)	Lokas et al. 2013
Rocky Flats	Soil	–	0.19 ± 0.12	–	Litaor 1995
Salzburg, Austria	Soil	–	0.44 0.13–1.68	0.057 0.019–0.232	Smcik et al. 2008
Rez, Czech Republic	Soil	2000	0.49 (0.31–0.86)	0.039 (0.024–0.059)	Holgye et al. 2004
Weapons-grade Pu (Thule accident)	Soil	2001	0.169 ± 0.005 (0.162–0.172)	0.0161 ± 0.0005 (0.0158–0.0167)	Eriksson et al. 2008
Weapons-grade Pu (Palomares accident)	Soil	1997	0.25C (0.15–0.35)	0.018 ± 0.003 (0.015–0.024)	Irlweck and Hmecek 1999
BOMARC	Soil	2003	0.19 ± 0.07 (0.15–0.24)	0.024 ± 0.009 0.017–0.033	Lee and Clark 2005
Vilnius, Lithuania	Air	2005–2006	0.40 (0.19–6.0)	0.033 (0.028–0.042)	Lujanienė et al. 2012a
Chernobyl fallout	Soil	1986	0.061–0.13	0.47–0.54	Aarkrog 1988b
Chernobyl fallout, Vilnius, Lithuania Chernobyl	Aerosol	1986	0.11 ± 0.05 (0.08–0.19)	0.48 ± 0.02 (0.44–0.50)	Lujanienė et al., 2009
Fangataufa Atoll	Soil	1997	0.05 ± 0.01 (0.037–0.077)	0.31 ± 0.09 (0.13–0.42)	Irlweck and Hmecek 1999
Mururoa Atoll	Soil	1997	0.017 ± 0.005 (0.009–0.027)	0.0054 ± 0.002 (0.004–0.009)	Irlweck and Hmecek 1999
Rice field, Japan	Soil	1957–1980	0.33 ± 0.03 (0.24–0.45)	0.30 ± 0.005 (0.012–0.039)	Yamamoto et al. 1983
Sodankyla, Finland	Aerosol	2007	–	0.045 (0.014–0.32)	Salminen and Paatero 2009
Semipalatinsk-STS Onsite	Soil	1994	0.015–0.447	0.0185–0.113	Yamamoto et al. 1996
Semipalatinsk-STS, near Kazakhstan	Soil	1994	0.016–0.040	0.11–0.35	Yamamoto et al. 1996
Thule	Sediment	1968	0.019 ± 0.006	0.05	Aarkrog et al. 1984

been measured in soils from Bikini Atoll (Muramatsu et al. 2001). However, fallout from Nagasaki, the Nevada Test Site, and the Semipalatinsk Test Site in the former Soviet Union, is characterized by lower  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios ranging from 0.03 to 0.08, with average values of 0.042, 0.035, and 0.036, respectively (Yamamoto et al. 1996). Therefore, WIPP soils do not appear to have been measurably affected by resuspension and transport of Gnome site soil, WIPP waste disposal operations, or the February 2014 accidental release, but reflect normal global fallout.

### Global redistribution of radionuclides in the atmosphere

Several studies have shown that resuspension is a dominant mechanism in maintaining residual concentrations of  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  in the post-fallout air. For example, concentrations in aerosols sampled near contaminated sites such as Rocky Flats, the Nevada Test site, the Marshall Islands (Shinn et al. 1997; Hulse et al. 1999), and in areas near the Semipalatinsk Test Site (Lehto et al. 2006) were elevated.

Resuspension studies performed with  $^{241}\text{Am}$  indicate that the activity to mass ratio is greatest for particles  $< 45\ \mu\text{m}$  in diameter (Burns et al. 1995). This is because fine soils, which have a higher surface area and capacity to sorb these radionuclides, have higher concentrations of  $^{241}\text{Am}$  than the remaining surface soil. These fine soils are also easily resuspended and transported by air, giving rise to a so-called enhancement factor. The enhancement factor for resuspended soil, in the inhalable fraction ( $< 7\ \mu\text{m}$ ), ranged from 3.7 to 32.5 at six sites at Taranaki in Southern Australia (Burns et al. 1995).

Shinonaga et al. (2014) reported long-distance transport of Fukushima-derived plutonium and uranium particles by wind. Environmental processes such as global dust storms, biomass burning, and/or wildfire have also been shown to play an important role in redistributing post-fallout radionuclides, including  $^{241}\text{Am}$ , in the atmosphere. Recent wildfire events of April and August 2015, in the Chernobyl exclusive zone, are estimated to have released about 12.5 TBq ( $10^{12}$  Bq) of radioactivity, mostly  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$ ,  $^{238}\text{Pu}$ , and  $^{241}\text{Am}$ , into the atmosphere (Evangelidou et al. 2016). These authors also concluded that during the fire events of August 2015,  $\sim 75\%$  of ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ) and  $59\%$  of ( $^{239+240}\text{Pu}$ ,  $^{238}\text{Pu}$ , and  $^{241}\text{Am}$ ) were exported from the Chernobyl exclusive zone and deposited mostly in Belarus, while during the spring fire event of 2016,  $\sim 93\%$  of the  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , and  $97\%$  of the  $^{239+240}\text{Pu}$ ,  $^{238}\text{Pu}$ , and  $^{241}\text{Am}$ , were transported to Eastern European countries. Wotawa et al. (2006) reported that radionuclides resuspended by wildfires can be transported over intercontinental distances. Lujanienė et al. (2006) attributed detection of an elevated level of  $^{137}\text{Cs}$  ( $253\ \mu\text{Bq}/\text{m}^3$ ) in air over Vilnius, Lithuania, to the transport of Chernobyl-derived cesium from the Chernobyl exclusion zone in Ukraine following the wildfire event of 1992.

Soil dust is a major component of aerosols in the global atmosphere and dust transport events that move African Sahara dust and Asian Kosa dust are known to play a major role in redistribution of radionuclides in the atmosphere. A significant increase in anthropogenic radionuclide concentrations in air has been observed during Saharan dust events in Europe, in the USA, and Asia (Menut et al. 2009). The  $^{239+240}\text{Pu}$  activity concentrations in Saharan dust samples vary between 0.68 and 0.98 Bq/kg (Pham et al. 2017), which is significantly higher than the 0.096 Bq/kg usually found in Saharan soil. Sequential extraction studies were performed on aerosols collected in Lithuania after dust storms in September 1992 carried radioactive aerosols to the region from contaminated areas of the Ukraine and Belarus. The fraction distribution of  $^{241}\text{Am}$  in the aerosol samples was  $\sim 18\%$  organically bound,  $\sim 10\%$  oxide-bound,  $\sim 36\%$  acid-soluble, and  $\sim 32\%$  residual (Lujanienė et al. 1999). A very small amount of americium was found in the more readily extractable “exchangeable and water soluble” and “specifically adsorbed” fractions.

The primary source of anthropogenic radionuclides in dust transported globally is surface soil contaminated by global fallout, and possibly by regional fallout from the accidents at Chernobyl and Fukushima. The activity and atom ratios of dust samples collected during several past dust events confirm that they originate from global fallout origin, and/or mixing with local resuspended soil particles during transport.

## Conclusion

The  $^{241}\text{Am}$  present in the environment today originated directly from nuclear reactors or indirectly from weapons testing. At nuclear facilities like the WIPP repository, being able to determine the origin of transuranium contaminants found in the environment is important for improving radiological protection programs and demonstrating regulatory compliance. Pre-operations measurements at the WIPP site established a background  $^{241}\text{Am}$  concentrations of  $0.027 \pm 0.11\ \mu\text{Bq}/\text{m}^3$  in aerosols and  $0.066 \pm 0.001\ \text{Bq}/\text{kg}$  in soil. The  $^{241}\text{Am}$  activities in aerosol and soil are determined to be not significantly different from those due to atmospheric global fallout. The Gnome site is a potential local source of contamination of WIPP environmental samples and has a characteristic isotopic signature. Long-term monitoring of  $^{241}\text{Am}$  concentrations show that, except for a brief detection of elevated americium and plutonium in nearby ambient air samplers during the 2014 accidental release from the repository, there is no evidence of contamination that can be attributed to operations at the WIPP. The source of  $^{241}\text{Am}$  at the site is mostly wind-borne redistribution of resuspended soils contaminated by weapons testing. During most years studied, the peaks in  $^{241}\text{Am}$  activities, like that of  $^{239+240}\text{Pu}$ , generally occurred in the March to June timeframe, which is when strong and gusty winds in the area frequently give rise to blowing dust. The isotopic signature of  $^{241}\text{Am}$  in the WIPP environment is mainly from three different sources with  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratios of  $\sim 0.37$  from global fallout,  $\sim 0.25$  as a result of Project Gnome fallout, and  $\sim 10$  due to the accidental radiation release in February 2014. Soil monitoring in the vicinity of the WIPP site suggests that the WIPP soils have received their plutonium and americium from global fallout during nuclear weapons testing and have not been measurably affected by resuspension and transport of Gnome site soils or by WIPP waste disposal operations. There is no reason to believe that WIPP is a source of transuranium contaminants in the environment that can be considered significant by any health-based standard.

**Acknowledgements** This research is supported by grant from US Department of Energy, Carlsbad Field Office of DOE through Grant No. DE-EM 0002423. Any opinions, findings and conclusions or

recommendations expressed in this material are those of the authors and do not necessarily reflect the views of the sponsors.

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