Contract No.:

This manuscript has been authored by Savannah River Nuclear Solutions (SRNS), LLC under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

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Plutonium-244 in the environment: Insights into plutonium's longest-lived isotope

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Highlights

- We report the first detailed long-term study of ^{244}Pu occurrence in the environment.
- Eleven year study of plutonium in terrestrial environmental samples from the Savannah River Site, USA.
- Significant ^{244}Pu was measured in all years.
- The 2003 sample contained 4423 fg of ^{244}Pu with a ^{244}Pu/^{239}Pu atom ratio of 0.70.

ABSRACT

This work describes the first detailed investigation of anthropogenic ^{244}Pu in the environment. Terrestrial environmental samples were taken at Savannah River Site, USA (SRS) over an eleven year period, from 2003 to 2014. Separation and purification of trace plutonium was carried out followed by three stage thermal ionization mass spectrometry (3STIMS) measurements for plutonium isotopic content and isotopic ratios. Significant ^{244}Pu was measured in all of the years sampled; and the highest values were observed in 2003. The ^{244}Pu content ranged from 31 to 4423 femtograms (fg = 10^{-15} g) in years 2006 and 2003 respectively. The ^{244}Pu/^{239}Pu atom ratios ranged from 0.0043 to 0.6981 in years 2009 and 2003 respectively.

Keywords
plutonium-244; high-flux reactors; nuclear and thermonuclear detonation; plutonium production; californium-252; three stage thermal ionization mass spectrometry

1. INTRODUCTION

1.1. Background

With a half-life of 81 million years, plutonium-244 is the longest-lived plutonium (Pu) isotope. Plutonium-244 formation is expected to occur in nature via cosmogenic processes, most notably in supernovae [1-3]. Although the neutron fluxes are insufficient for ^{244}Pu to be produced naturally in uranium ores, this isotope has been observed in the Earth's crust in very small quantities, e.g., in the rare-earth mineral bastnäsite (Hoffman et al., 1971; Lachner et al., 2012). As such, ^{244}Pu may be the heaviest relic on Earth of the last supernova(e) that occurred before the formation of the Solar System (Alexander et al., 1971; Lachner et al., 2012).

A limiting factor hampering anthropogenic ^{244}Pu production is that, because neutron capture by ^{242}Pu produces ^{243}Pu, timely neutron capture must occur by ^{243}Pu before it beta decays, with a roughly 5 hour half-life, to americium-243. Man-made creation of ^{244}Pu therefore can only occur under extreme conditions, and typically via one of two pathways: In a high-flux reactor ($\geq 5 \times 10^{15}$ neutrons per square centimeter per second) with a heavy isotope target - conditions not typically encountered in commercial or weapons production reactors - or almost instantaneously during a nuclear or thermonuclear weapon detonation.

Whether originating from natural or anthropogenic provenances ^{244}Pu is typically found in the terrestrial environment in relatively small amounts, and, correspondingly, studies of ^{244}Pu occurrence in the environment are sparse. Indeed the first (and only) atmospheric global fallout ratio for ^{244}Pu (^{244}Pu/^{239}Pu = 5.7×10^{-5}) has only recently been put forth (Steier et al., 2013).

In addition to its relative scarcity, analysis of ^{244}Pu by conventional means, i.e., alpha spectrometry and mass spectrometry, is a challenge. The radiological properties of plutonium isotopes 236 – 244 are shown in **Table 1**. Owing to its relatively small specific activity, ^{244}Pu measurement by alpha spectrometry is generally impractical. Additionally, ^{244}Pu is commonly used as an internal tracer for plutonium analysis by mass

spectrometry. In such cases any ^{244}Pu in the sample itself is assumed to be negligible and cannot be measured accurately.

Table 1. Summary of the radiological properties of the most relevant plutonium isotopes (Baum, 2009; Physics, 2005).

	Pu-236	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Pu-244
Half-life (in years)	2.9	87.74	24,110	6537	14.4	375,000	8.11 E7
Specific activity (curies/gram)	540	17.3	0.063	0.23	104	0.004	1.9E-5
Principal decay mode	alpha	alpha	alpha	alpha (some spontaneous fission)	beta	alpha	alpha
Decay energy (MeV)	5.768	5.593	5.244	5.255	0.021	4.983	4.6
Radiological hazards	alpha, weak gamma	alpha, weak gamma	alpha, weak gamma	alpha, weak gamma	beta, weak gamma	alpha, weak gamma	alpha, weak gamma

1.2. Need for Plutonium-244

The world's stock of ^{244}Pu is rapidly depleting and, correspondingly, it is becoming an increasingly precious isotope. This is a concern primarily because, as mentioned previously, ^{244}Pu is a common internal standard used in mass spectrometry plutonium analysis. Indeed this shortage has caused a major plutonium-244 supplier, New Brunswick Laboratories (NBL), to hold the remainder of its supply. To meet the growing need for this precious commodity, recent interest has been sparked to harvest the isotope from legacy materials, for example, from targets that were irradiated during the californium-252 production campaigns at SRS decades ago. A specific example of such a material is the Mark-18 (Mk-18A) target. From August 1969 until November 1970, eighty-six Mk-18A targets were irradiated in a high-neutron-flux mode in the 2000 megawatt-thermal (MWt) K-Reactor at SRS (Goldberg, 2001; Robinson, 2013). This intense exposure resulted in Mk-18A targets with very unique contents. Upon removal from the reactor, 21 targets were processed at Oak Ridge National Laboratory (ORNL) in

1972–1973 to recover ^{252}Cf, heavy curium, and plutonium. The plutonium fraction, which was rich in ^{244}Pu, was electromagnetically separated in the calutrons at ORNL to produce gram quantities of 98–99% ^{244}Pu. These high-purity samples of ^{244}Pu were portioned out to scientists for basic research and for safeguards programs. The remaining Mk-18A targets are expected to contain 21 grams of plutonium-244 which represents greater than 90 % of the world's inventory of this rare material. These remaining Mk-18A targets are currently being stored at SRS. Researchers at Savannah River National Laboratory (SRNL) and ORNL have recently proposed to process these remaining targets to recover plutonium-244, heavy curium, and californium-252 at SRS (REF).

1.3. Historical studies of Plutonium-244

At present the only detailed studies of ^{244}Pu in the environment are related to the thermonuclear weapons testing in the northern Marshall Islands. Between 1946 and 1958, the United States conducted 43 thermonuclear detonation tests at Enewetak and Bikini atolls. These tiny land masses provide some of the few locations where measurable quantities of heavy elements and isotopes can be sampled. For example, an examination of a sample related to the 10.4 megaton Ivy Mike test (Bliss, 1982) detonated on November 1, 1952, lead to the discovery of ^{244}Pu, as well as elements Einsteinium (Es) and Fermium (Fm) (Ghiorso et al., 1955). A follow up study (Diamond et al., 1960) examined debris from the Mike test fallout and provided equally fruitful results. Diamond et al. noted that the Mike explosion produced, via neutron capture by ^{238}U, uranium isotopes ^{239}U to ^{255}U (inclusively). Measurements of the beta-decay products of these unstable isotopes resulted in the identification of a plethora of heavy elements and associated isotopes produced from the Mike test (**Fig. 1**), including a ^{244}Pu/^{239}Pu atom ratio of ~ 0.003.

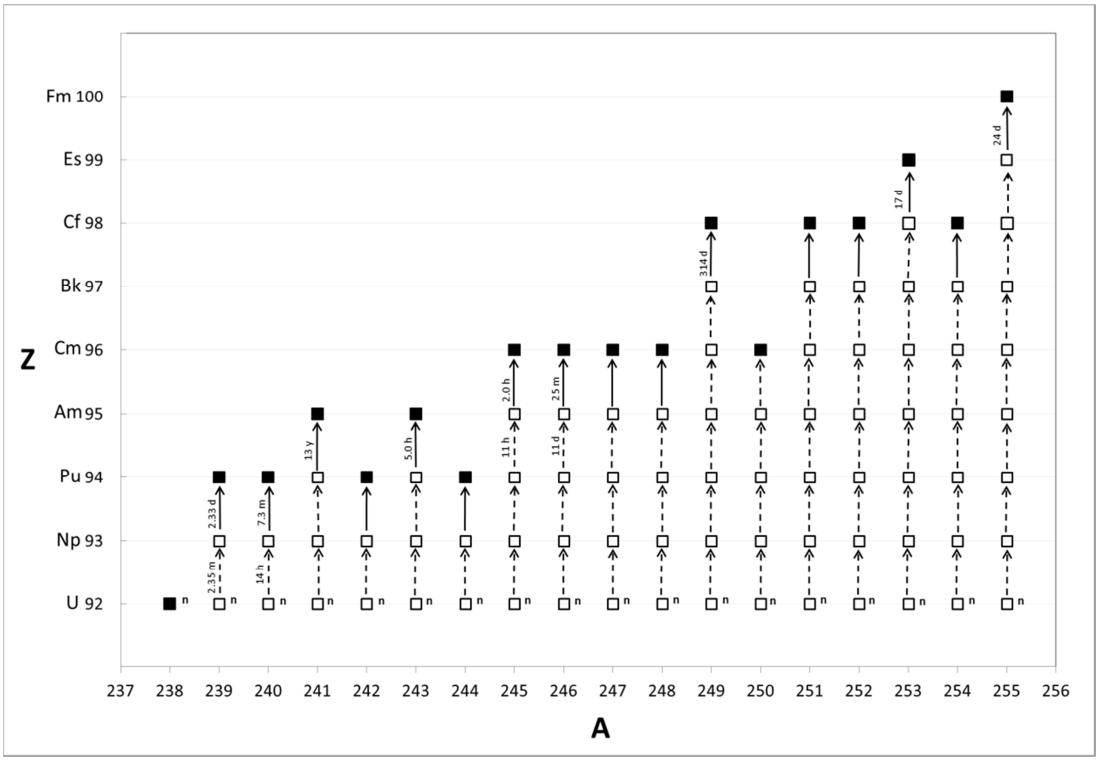

Fig. 1. Production of uranium isotopes in the November 1952 Mike thermonuclear event and the associated beta decay products. Modified from Diamond et al, 1960. ▫ beta unstable nuclide. ▪ beta stable nuclide.

In a more recent study (Lachner et al., 2010), soils were collected from the Bikini atoll and $^{244}Pu/^{239}Pu$ ratios ranging from roughly $2 – 5 \times 10^{-4}$. In another recent study (Hamilton et al., 2009), soil sample collected from Enewetak atoll exhibited a $^{244}Pu/^{239}Pu$ atom ratio of 3.2×10^{-5}.

1.4. Plutonium-244 production at the Savannah River Site (SRS)

The Savannah River Site (SRS), formerly the Savannah River Plant (SRP) began operation in the early 1950's. The site, located close to the Savannah River in South Carolina (Figure 1), encompasses roughly 800 square kilometers. Its primary function was to produce special nuclear materials for national defense, specifically weapons-grade plutonium (Pu) and tritium (^{3}H) required for thermonuclear devices (Bebbington, 1990). Over its history the site produced metric tons of plutonium comprising Pu isotopes with masses 238 through 244.

The highly versatile reactors at SRS provided a unique breeding ground for ^{244}Pu as this isotope was produced in SRS reactors as a byproduct of the californium-252 production campaigns. Notably, during the aforementioned 1969-1970 period , the most ^{252}Cf ever made, some 2.1 g, was produced in the SRS K-Reactor (Boswell, 2000). To accomplish this feat, targets containing primarily Pu-242 were introduced into the high-flux reactor. In addition to plutonium-244, many heavier isotopes were also produced during this process, including the heavier curium isotopes, e.g., 245 – 248.

Owing to its unique plutonium production history, SRS offers a unique environment for detailed study of plutonium fate and transport, and specifically ^{244}Pu. We recently reported findings of a long-term study of plutonium detailing sampling, radiochemical processing, and analysis of plutonium in environmental samples from SRS (Armstrong et al., 2015). This previous study reported ^{238}Pu - ^{242}Pu (inclusively) data for terrestrial environmental samples collected over a ten year timespan. This current contribution expands on this work reporting ^{244}Pu results over an eleven year timespan.

In contrast to the aforementioned studies of anthropogenic ^{244}Pu related to thermonuclear weapon testing (Bliss, 1982; Diamond et al., 1960; Hamilton et al., 2009; Lachner et al., 2010), this current study reports the occurrence of reactor-produced ^{244}Pu in the terrestrial environment. Collectively, these studies afford unique insights into the fate and transport of Pu-244. For example, if the proposed processing of the aforementioned Mark-18A targets moves forward, a background assessment of Pu-244 levels at SRS will be necessary. More broadly, these investigations augment the global database of plutonium occurrence in the environment thereby enriching the state of knowledge of plutonium fate and transport in the environment. Insights gained from Pu-244 studies are therefore of interest from a variety of disciplines, including fundamental science (astrology, cosmology, etc.), national security, and nuclear nonproliferation.

2. EXPERIMENTAL

2.1. Methodology

This study is an extension of a previous investigation (Armstrong et al., 2015). Briefly, the environmental samples in this study consist of materials collected from the bottom of

visitors' footwear via a mechanical shoe brush (www.liberty-ind.com) before entering the clean laboratories at Savannah River National Laboratory (SRNL). The materials collected consist of mineral fragments, vegetation debris, soil, etc. This mixture of materials collected and transported from throughout the SRNL A-Area, offer a unique opportunity to collect bulk samples representative of the nearby environment. Samples are collected periodically (typically once or twice per year) and processed via the SRNL radiochemical processing procedure specifically designed for trace level plutonium purification and separation (Armstrong et al., 2015). The separated and purified plutonium samples are analyzed first by alpha spectrometry and then by mass spectrometry for total plutonium and plutonium isotopic composition determination.

2.2. Sample Preparation, alpha spectrometry, and mass spectrometry

All glass, quartz, and Teflon materials are pretreated (leached) by refluxing for several hours at 200 ^{o}C in 8 M nitric acid. Type 1 (18 MΩ) water and high purity semiconductor grade acids are used exclusively this study.

Experimental details related to this study have been described previously (Armstrong et al., 2015). Briefly, upon extraction from the shoe brushes, samples are mixed manually and approximately 100 grams is subsampled and weighed out. All samples are ashed for at least eight hours in a furnace at 600 ^{o}C to destroy organic components in the matrix. The samples are subsequently digested and centrifuged in accordance with a well-established method (Greiling and Lieser, 1984) and the solid residue is archived for periodic monitoring of refractory Pu. Half of the sample is transferred by weight to a leached pyrex beaker and spiked with ^{242}Pu (15 – 20 pg from NIST SRM 4334G). The remaining solution is archived. After an oxidation state (to Pu(IV)) adjustment and co-precipitation step trace plutonium purification is performed according to an original methodology (Crouch and Cook, 1956) whereby samples are loaded and eluted through successive columns of AG1X8 resin (Eichrom) and AG1X4 (Eichrom) ion exchange resins.

2.3. Three stage thermal ionization mass spectrometry (3STIMS)

Plutonium measurements were conducted with a 1960's KAPL (Knolls Atomic Power Laboratory) design Three Stage Thermal Ionization Mass Spectrometer (3STIMS) fabricated in-house in the 1970's. The instrument uses three 90^0 x 30.5 cm sectors arranged in magnetic/magnetic/electrostatic order (momentum/momentum/energy filter order) with a single ion counting detector. This arrangement allows for measurement of very high ratios between adjacent ion masses, of order 108, but the instrument must be sequentially tuned from mass to mass to collect ions at each isotope, less efficient than an instrument with a detector per ion beam. It routinely analyzes nanogram mass U samples and picogram mass Pu samples. Purified samples are loaded onto anion exchange resin beads, which are then loaded by hand onto high-purity rhenium filaments and placed in the source region of the mass spectrometer for thermal ionization.

2.4. 3STIMS measurements

Isotopic content measurements of ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{244}Pu together with atom ratios of ^{240}Pu/^{239}Pu, ^{241}Pu/^{239}Pu, ^{242}Pu/^{239}Pu, and ^{244}Pu/^{239}Pu were carried out by three-stage thermal ionization mass spectrometry (3STIMS). These data are corrected for impurities in the standards. Reagent blank and bench standard samples were run with every sample set. All 3STIMS data are mass bias corrected. Stringent quality assurance and quality control measures are routinely undertaken with the 3STIMS instrument. A two-sigma relative standard deviation (RSD) of approximately 10 % is expected for measurements of Pu in the 10 fg (fg = 10^{-15} g) range. In addition to employing internal standards, an external standard (certified ratio standard: New Brunswick CRM-128) consisting of 10 – 30 pg total Pu of a 1:1 ^{239}Pu:^{242}Pu with trace ^{238}Pu, ^{240}Pu and ^{241}Pu is ran with every sample set. For Pu isotopic abundances excellent accuracy is observed with this instrument. For example, when compared to the certified (CRM-128) value, the 239/242, 240/242, and 241/242 ratios are routinely within analytical error (typically 0.5 % or better).

3. RESULTS AND DISCUSSION

3.1 Plutonium isotopic content results

For years in which a ^{242}Pu internal standard (tracer) was added during radiochemical processing (2003, 2004, 2005, 2006, 2009, and 2011), the plutonium isotopic content was quantified for isotopes ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{244}Pu by three stage thermal ionization mass spectrometry (3STIMS). These isotopic mass data, in femtograms, are shown in **Fig. 2** on a logarithmic scale, and reported in **Table 2.** Mass results of isotopes $^{239\text{-}242}$Pu are included in **Fig. 2** and **Table 2** for comparative purposes; but the focus of this discussion is restricted to ^{244}Pu. Significant amounts of ^{244}Pu were observed in all years, with amounts ranging from 31 to 4423 fg in years 2006 and 2003 respectively.

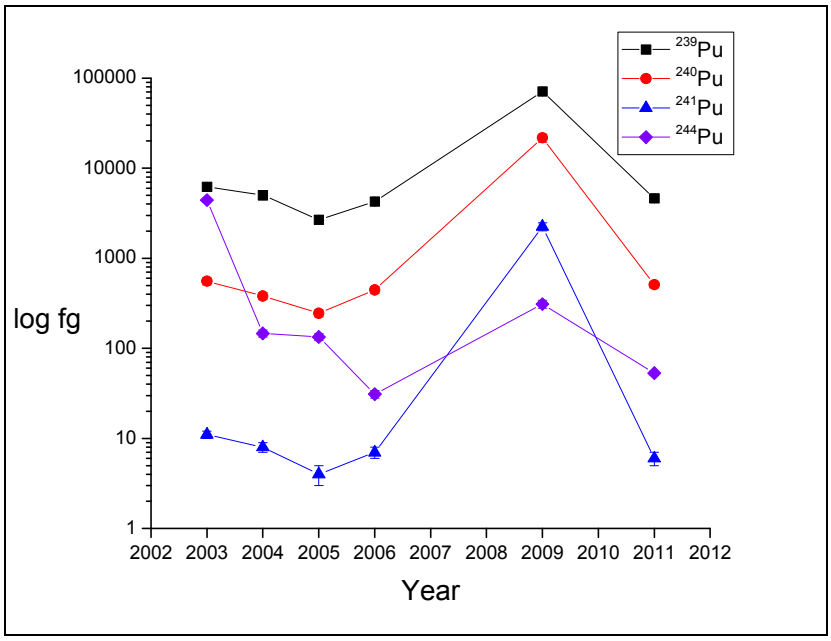

Fig. 2. Plutonium isotopic content from three stage thermal ionization mass spectrometry (3STIMS) measurements (logarithmic scale) in femtograms with two-sigma error. Data are reported for sample years in which a ^{242}Pu internal standard was added to the samples.

The 2009 sample year featured anomalously high collections of ^{239}Pu through ^{241}Pu. Although a significant amount of ^{244}Pu was also measured in 2009 (310 fg) it was well

below the highest collection year in 2003. The marked elevation in ^{239}Pu, and particularly in ^{240}Pu and ^{241}Pu, was most likely due to plutonium oxide processing activities that occurred during this time frame in an adjacent laboratory in A-Area at SRNL (Armstrong et al., 2015). These materials contained relatively high ^{240}Pu and ^{241}Pu; and it is conceivable that a small contribution was tracked into our laboratory and picked up by the shoe brushes.

Table 2. Plutonium isotopic content from three stage thermal ionization mass spectrometry (3STIMS) measurements in femtograms with two-sigma error. Data are reported for sample years in which a ^{242}Pu internal standard was added to the samples.

Year	^{239}Pu	^{240}Pu	^{241}Pu	^{244}Pu
2003	6206 ± 561	557 ± 50	11 ± 1	4423 ± 200
2004	5009 ± 452	381 ± 34	8 ± 1	146 ± 13
2005	2684 ± 242	245 ± 22	4 ± 1	134 ± 12
2006	4262 ± 385	445 ± 40	7 ± 1	31 ± 3
2009	71288 ± 7190	21724 ± 2191	2251 ± 227	310 ± 31
2011	4634 ± 343	510 ± 39	6 ± 1	53 ± 3

3.2 Plutonium atom ratio results

Atom ratio data for ^{240}Pu/^{239}Pu, ^{241}Pu/^{239}Pu, ^{242}Pu/^{239}Pu, and ^{244}Pu/^{239}Pu for this eleven year study are reported in **Table 3**. The isotopic ratios for isotopes 239 – 242 from this sample set, depicted as the shaded region in **Table 3** and included for comparison, have been reported previously in our initial report (Armstrong et al., 2015). With the exception of 2009 in which a higher burnup signature is observed (see ^{240}Pu/^{239}Pu and ^{241}Pu/^{239}Pu in **Table 3**), these data generally appear to be representative of a variable admixture of both fallout and weapons-grade plutonium, with several ratios approaching those of weapons-grade Pu. Relative to atmospheric fallout, elevated ^{244}Pu/^{242}Pu atom ratios were measured in every sample year. As also reported in **Table 2**, the 2003 sample year featured the most pronounced ^{244}Pu enrichment with a ^{244}Pu/^{239}Pu atom ratio of approximately 0.70.

Table 3. Three stage thermal ionization mass spectrometry (3STIMS) plutonium atom ratios for SRS environmental samples spanning a ten year period with two-sigma error. Shaded values were initially reported in Armstrong et al., 2015; [d]Zheng et al., 2013, [e]Oughton et al., 2000, and [f]Steir et al., 2013. (Steier et al., 2013).

	3STIMS (atom ratios)			
Year	$^{240}Pu/^{239}Pu$[a]	$^{241}Pu/^{239}Pu$[b]	$^{242}Pu/^{239}Pu$[c]	$^{244}Pu/^{239}Pu$
2003	0.0894 ± 0.0007	0.00189 ± 0.00009	NA	0.6981 ± 0.0034
2004	0.0757 ± 0.0004	0.00166 ± 0.00006	NA	0.0286 ± 0.0002
2005	0.0908 ± 0.0005	0.00158 ± 0.00006	NA	0.0488 ± 0.0004
2006	0.1040 ± 0.0005	0.00155 ± 0.00005	NA	0.0072 ± 0.0001
2007	0.0945 ± 0.0001	0.00148 ± 0.00077	0.01875 ± 0.00014	0.0215 ± 0.0001
2008	0.0952 ± 0.0003	0.00155 ± 0.00002	0.01968 ± 0.00008	0.0224 ± 0.0001
2009	0.3035 ± 0.0015	0.03131 ± 0.00024	NA	0.0043 ± 0.0001
2010	0.1083 ± 0.0003	0.00129 ± 0.00003	0.01851 ± 0.00012	0.0249 ± 0.0004
2011	0.1097 ± 0.0024	0.00132 ± 0.00026	NA	0.0112 ± 0.0007
2012	0.1128 ± 0.0016	0.00178 ± 0.00011	0.01733 ± 0.00051	0.0152 ± 0.0003
2013	0.1322 ± 0.0011	0.00124 ± 0.00010	0.04273 ± 0.00055	0.0203 ± 0.0003
2014	0.1516 ± 0.0026	0.00106 ± 0.00010	0.02778 ± 0.00070	0.0026 ± 0.0002
Fallout	0.180 ± 0.007[d]	0.00194 ± 0 .00014[e]	0.0039 ± 0.0007[e]	5.7E-05[f]

The $^{244}Pu/^{239}Pu$ atom ratios reported in Table 3 are significantly higher than the values reported from the recent Marshall Island studies (Lachner et al., 2010; Hamilton et al., 2009). Moreover, with the exception of the 2003 sample year, all of the samples exhibit $^{244}Pu/^{239}Pu$ atom ratios higher those originating from the Mike thermonuclear detonation test (~0.003; Diamond et al., 1960). These quantities of ^{244}Pu are representative of a sample set that is unique to the SRS environment, where gram quantities of ^{244}Pu were made in the specially-designed high-flux reactors that once operated at SRS. Resuspension of these legacy materials resulted in a general dispersion of ^{244}Pu containing solid phases such that small quantities are readily quantifiable.

4. CONCLUSIONS

The work presented herein details the findings of an eleven year study of plutonium in terrestrial environmental samples from the Savannah River Site, USA. This long-term inquiry provides the first detailed study of ^{244}Pu occurrence in the environment. The following conclusions are drawn. Although the highest amount of ^{244}Pu originated from the 2003 year, significant ^{244}Pu was measured in all years. The ^{244}Pu content ranged from 31 to 4423 fg in years 2006 and 2003 respectively. The ^{244}Pu/^{239}Pu atom ratios ranged from 0.0043 to 0.6981 in years 2009 and 2003 respectively. The plutonium signatures obtained in this study reflect a provenance of plutonium originating from the unique high-flux reactors at SRS where ^{242}Pu was target material primarily for ^{252}Cf production. Due to the paucity of ^{244}Pu data available, provide unique insights that may be helpful in advancing both remediation and post-detonation assessment strategies.

5. ACKNOWLEDGEMENTS

The authors gratefully acknowledge Sheila Smalley for providing technical assistance, particularly in performing the radiochemical separations throughout her many years of service at Savannah River National Laboratory. This document was prepared in conjunction with work accomplished under Contract no. DE-AC09-08SR22470 with the U.S. Department of Energy.

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