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Ultra-low-level measurements of airborne fission products from the Fukushima Daiichi reactor accident using high volume collection systems at Savannah River National Laboratory

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Abstract

Ultra-low-level measurements of radionuclides in air have been conducted at the Savannah River National Laboratory (SRNL) to determine the atmospheric concentration of fission products released following the Fukushima Daiichi reactor accident on 11 March 2011. Air filter samples were acquired from two high-volume collection systems (a traditional filter-based system and an electrostatic precipitator-based system) to monitor airborne radionuclide concentrations in the period covering from 2 weeks to 3 years after the disaster. The world-wide spread of low-level concentrations of airborne fission products from the Fukushima event provided a unique opportunity to demonstrate SRNL's electrostatic particle collection technology and other improvements in environmental monitoring developed at the Savannah River Site (SRS). Detecting and analyzing the release allowed a comprehensive test of SRS systems for monitoring environmental radioactivity. Gamma-ray-emitting fission products (^{131,132}I, ^{134,136,137}Cs, and ^{129,132}Te) and cosmogenic isotopes (^{7}Be and ^{22}Na) in air were detected and quantified by high-resolution gamma-ray spectroscopy at concentrations as low as 0.07 μBq per standard cubic meter (SCM) (50 mBq total ^{137}Cs), while plutonium content was quantified by thermal ionization mass spectrometry (TIMS) at concentrations as low as 6.5×10^{-21} g/SCM (3.0 femtograms $^{239+240}$Pu). Isotope concentrations measured at SRNL from gamma-ray spectroscopy were compared to independent measurements from Chapel Hill, NC, located approximately 370 km (230 mi) NE of SRNL. Meteorological modeling was also used to predict radionuclide

transport from the location of release to both measurement locations.

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1. Introduction

Air particulate samples have been collected and analyzed at the Savannah River Site (SRS) since the early 1950s and radio-analytical techniques have been developed to distinguish SRS produced radionuclides, natural background, and fallout from historic and present-day nuclear testing. A low-level radiochemistry group was formed at the SRS to develop methodology and support these monitoring efforts as part of the Analytical Chemistry Division. This group evolved into the Nuclear Nonproliferation Division of Savannah River National Laboratory (SRNL). Active environmental monitoring continues through the present day, providing an extensive dataset for measuring the environmental impact of continuing SRS and SRNL operations as well as providing important tools for the broader nonproliferation community.

The reactor accident at Fukushima Daiichi in March 2011 presented a unique opportunity to integrate and test SRNL's ultra-low level air particulate sampling and analysis technology during a global radiological event. The primary goals of the present study were to determine the radionuclides observable in air in the Southeastern United States from the Fukushima release, to quantify their respective activities and practical limits of detection, and to measure their persistence. Secondary goals were to compare the relative levels of ^{131}I (a volatile fission product that often makes up a large percentage of activity initially released in reactor accidents) measured to SRNL model predictions and to provide data that will be useful to others in

improving radionuclide atmospheric transport modeling more generally. The first measurements of the airborne activity concentrations of transported fission products released during the Fukushima Daiichi reactor accident were begun at SRNL on 23 March 2011, 12 days after the disaster and before the peak activity of the plume was predicted to reach the East Coast of the United States. Two different air sampler systems were used to acquire measurements, a high-flow-rate filter-based system and a high-volume electrostatic precipitator system. The sensitivity of each system to atmospheric radionuclides was increased by gamma-ray counting of samples in SRNL's ultra-low-background Underground Counting Facility (UCF) (Winn et al., 1988).

2. Materials and Methods

2.1. Air Sampler Systems

A high-flow-rate air sampling system, installed on SRNL's Tracking Radioactive Airborne Contamination (TRAC) vehicle (which was designed for emergency response and use at remote research locations) took single-day measurements, with collection times between one and a half to six hours, on nine separate days between 25 March and 28 April 2011. The air sampling system installed in the rear of the TRAC vehicle consists of a 5 hp fan motor which draws outdoor air through a 24" × 24" (0.37 m^2) Filtrete^{TM} filter (3M Corporation) positioned at the level of the vehicle roof (~3 m above the ground) at an average flow rate of approximately 2200 standard cubic meters (SCM) per hour (see **Fig. 1**). The total air flow volume for each filter sample was measured with a calibrated mass flow sensor and totalizer (Model 502 In-Line Thermal Mass Flow Element from Kurz Instruments, Inc.), and the uncertainty in the resultant total air volume was ± 0.5 standard cubic feet (SCF) (0.014 SCM). At the measured flow rate, the collection efficiency of the filter was determined to be 80 ± 2% as measured by ^{7}Be trapped by stacked filters; the collection efficiency for other measured radionuclides was considered to

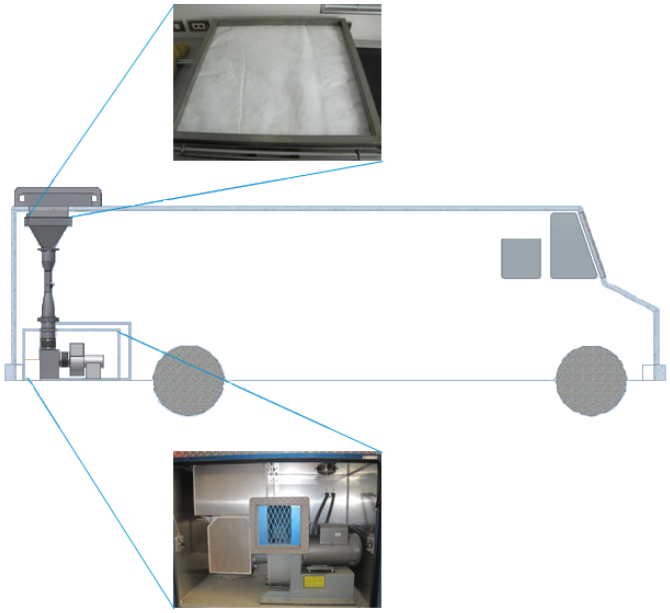

Figure 1. Diagram of the TRAC vehicle and associated air sampling system used for high-flow-rate, single-day collections.

be the same for the purposes of the present study. The collection location was about 1 km from the sand filter stack that filters radionuclide releases from research activities at SRNL; these radionuclide releases are monitored and did not impact measurements of radionuclides from the Fukushima event. After sample collection, each TRAC filter was folded and rolled into a 10 cm diameter by 12 cm high cylindrical geometry and placed within a 1 L polypropylene vial for gamma-ray counting.

The second air sampling system used for measurements consisted of a set of identical, portable electrostatic precipitators developed at SRNL and based on modified commercial technology. The samplers use an applied high voltage to fix particles to a removable precipitator substrate consisting of thin, high-purity aluminum plates. Each sampler has a flow rate of approximately 600 SCM/hr and can run continuously for up to 12 weeks. These units were used to perform longer duration ground (and later roof-top) level measurements (from 2 weeks to several months collection time) beginning 23 March 2011 and ending 16 July 2014, giving higher sensitivity for detection of trace-level fission products and cosmogenic radionuclides. The long, high-volume runs with the precipitators increased the total volume of air sampled and thus the sensitivity by an order of magnitude over the TRAC system. The flow rate was calibrated with a balometer, which measures the flow velocity through a defined cross-sectional area. The uncertainty in the total

volume of air sampled per run was ± 10%. The collection efficiency of the precipitators is presumed to be high, but the exact value is unknown. All electrostatic precipitator data presented in this paper gives the measured activity concentrations uncorrected for collection efficiency. The first set of measurements from 23 March 2011 to 30 Apr 2012 were taken with a wood-shielded electrostatic precipitator unit placed 1.8 m above ground level and about 100 meters downwind from the SRNL sand filter stack. The second electrostatic precipitator unit, shielded with metal, was used to perform roof-top level measurements (approximately 10 m above ground) on a nearby building about 50 meters downwind of the stack beginning on 17 Apr 2012. During mid to late 2011, the Honey Prairie Fire in the Okefenokee Swamp of southern Georgia (US) produced considerable amounts of soot at the SRS, which was collected by the electrostatic precipitator used during ground-level sampling, but the soot did not appear to affect the radionuclide measurements. After sampling runs, particulates were removed from the plates and suspended in aqueous solution by ultrasonic cleaning in hot water. The suspension was then evaporated to a volume of between 100 and 200 cm^3 (varied by sample) and placed in a 6 cm inner diameter (ID) by 8 cm high cylindrical polypropylene vial for gamma-ray counting.

2.2. Gamma-ray Counting and TIMS Measurements

Sample vials were cleaned with polypropylene wipes saturated with a mixture of 70% isopropyl alcohol and 30% de-ionized water to eliminate any external contamination. The vials were then placed within sealed plastic bags and counted directly on top of a high-efficiency, high-purity germanium (HPGe) detector with a J-type cryostat in the SRNL UCF. Several different detectors having relative efficiencies between 100 and 150% (i.e., compared to the standard reference efficiency of the 1332 keV gamma ray from a ^{60}Co source placed 25 cm from the front face of a 3"diameter × 3" thick NaI detector) were used for activity measurements. The

TRAC samples were all measured on a 150% p-type HPGe detector for counting times varying from 1 to 6 days, depending upon the activity level. The electrostatic precipitator samples were measured on one of four different detectors for counting times between 1 and 7 days. All sample activities were corrected for the decay of observed radionuclides during measurements. TRAC samples were also corrected for decay occurring between the sample acquisition date and date of measurement. Electrostatic precipitator samples were decay corrected to the median date and time for the sample collection period. Detector efficiencies were calibrated with gravimetrically prepared solutions of mixed radionuclide standards from Eckert & Zeigler Analytics traceable to the National Institute of Standards and Technology or the Deutschen Kalibrierdienst Kalibrierlaboratorium. ORTEC's Maestro® software was used for data acquisition.

All gamma-ray measurements were conducted in SRNL's UCF. The UCF consists of a 3.0 m × 4.3 m × 2.4 m counting chamber with 10.2 cm thick walls made from pre-WWII naval steel plate and placed 14.3 m underground. The room is additionally shielded by a minimum of 1.2 m of specular hematite on 5 sides as well as overhead layers of borated concrete and clay, giving total overburden shielding of 31.7 meters water equivalent (mwe). Individual detectors within the UCF are shielded with low-background lead. With this facility, ^{137}Cs can be detected at levels as low as 0.01 μBq/SCM. This high sensitivity has allowed us to make continuous measurements of the background levels of ^{137}Cs from global fallout as well as ^{22}Na produced by cosmic ray spallation, which local activity concentrations are on the order of 0.1 and 0.5 μBq/SCM, respectively. The typical counting times for samples measured in the UCF range from several days to 1 week. A cleanroom environment is maintained within the UCF and adjacent access areas (including the mass spectrometry laboratory where TIMS measurements were conducted), allowing for contamination-free sample preparation and processing. The facility is accredited for

gamma analysis under IEC/ISO 17025 and regularly participates in national and international quality assurance programs.

After gamma-ray measurements were completed on the electrostatic precipitator samples, plutonium fractions were chemically separated and purified from each sample and the isotopic composition subsequently measured on a three-stage thermal ionization mass spectrometer (3STIMS) using standard actinide radiochemistry. Additional details on radiochemical separation and analysis of plutonium-containing samples at SRNL can be found in Armstrong et al., 2015.

2.3. Atmospheric Transport Modeling

For the case of the Fukushima events, a Global Eulerian Model (GEM) (Draxler, 2007), which is a global version of the Hybrid Single-Particle Lagrangian Trajectory model (HYSPLIT) (Draxler, 1998), was used to predict the transport of radioactive aerosols to measurement locations at SRNL (33.34° N, 81.74° W) and the University of North Carolina at Chapel Hill (35.91° N , 79.05° W); the latter location was chosen as measurements of Fukushima releases were also conducted there and it is in the same geographic region as SRNL (MacMullin, et al., 2012). The GEM model used a simplified configuration to simulate global transport and dispersion of ^{131}I from Fukushima and assess when a radioactive plume might reach SRNL. Other studies using more sophisticated methods were also consulted to look at transport of radionuclides from Fukushima at local, regional, and global scales (Stohl, et al., 2012; Draxler et al., 2015; Saito et al., 2015; Solazzo and Galmarini, 2015). The simplified initial analysis was performed without detailed knowledge of the source terms.

An instantaneous unit release from Fukushima (37.42° N, 141.03° E) was assumed to occur for each day starting at 00:00 UTC on 12 March to 15 March 2011, which covers the release of ^{131}I emitted over the first 4 days of the event. Input meteorology at 1-degree resolution (~100 km)

using the Global Data Assimilation System (GDAS) (Kanamitsu, 1989; Kleist et al., 2009) was used to transport the material throughout the atmosphere. Activity concentrations are also generated on a grid with 1-degree spacing. The simulations were allowed to run for over one month, and output was generated once per day. An example showing global transport is provided in **Fig. 2**, where emissions from Fukushima (red dot) were directed to the east and over the Pacific Ocean before reaching the mainland United States. **Figure 2** shows the plume on 23 March 2011 at roughly the time when elevated airborne concentrations of radionuclides in the Southeastern United States were first being detected.

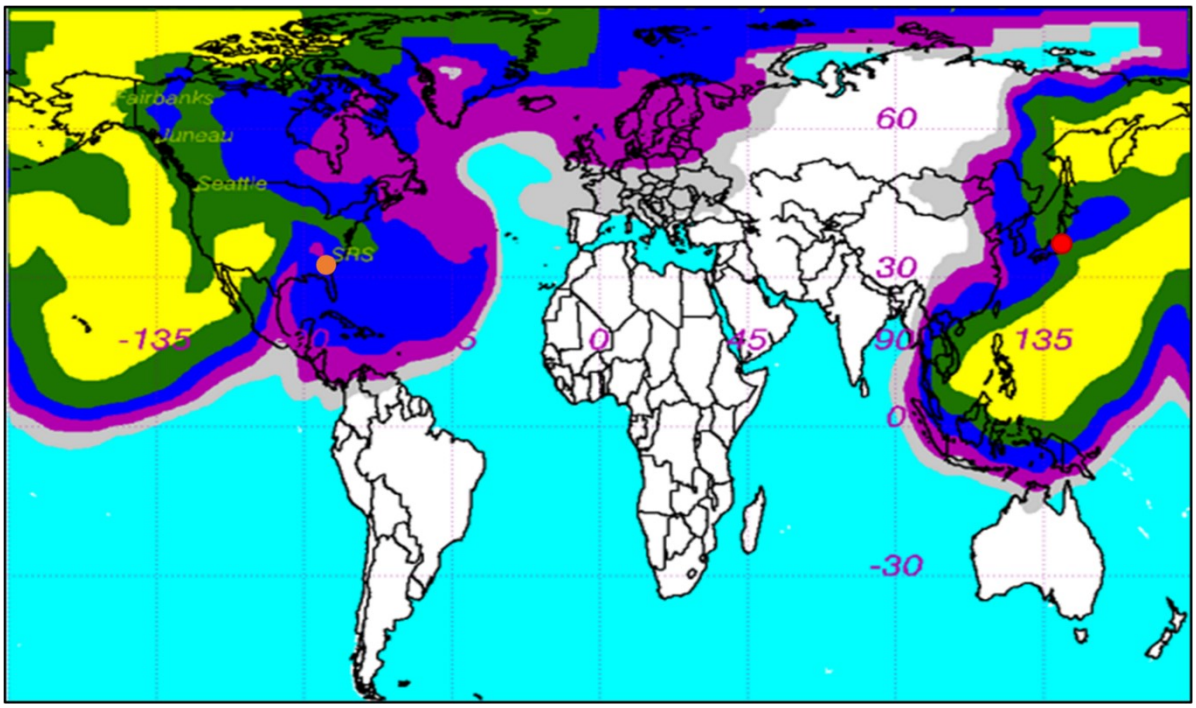

Figure 2. Simulated ^{131}I concentration using GEM valid at 00:00 UTC, 23 March 2011 for an assumed instantaneous unit release from Fukushima (large red dot) on 13 March 2011. Contour shading (yellow, green, blue, purple, gray) indicates descending concentration levels by order of magnitude where yellow denotes values between 10^{-7} and 10^{-6} units/SCM and gray denotes values between 10^{-11} and 10^{-10} units/SCM.

Standard bilinear interpolation of activity concentrations at SRNL and Chapel Hill, North Carolina from the 1-degree resolution output were created based on the different source release dates. Although particulates such as ^{131}I and ^{137}Cs will react with the environment (i.e., surface dry deposition or wet deposition due to precipitation scavenging), these loss effects were not

considered due to high uncertainty in the source conditions. Thus, the time series activity concentrations generated by the model are likely more representative of transport of noble gas releases such as ^{133}Xe which will not interact with the environment. The aim of the modeling was to provide an indication of when a radioactive plume might reach the Southeastern United States. Activity concentrations at both SRNL and Chapel Hill were produced for only the 15 March 2011 release time since this date corresponded to the peak ^{131}I release rate.

3. Results and Discussion

3.1. TRAC

Figure 3 shows the gamma-ray spectrum between 180 and 980 keV for the TRAC data acquired on 29 March 2011, on which date the peak activity levels for all fission radionuclides except ^{131}I (which peaked on 1 April 2011) were observed, and several months later on 9 June 2011. Gamma-rays corresponding to multiple fission products including ^{131}I, ^{129,129m}Te, ^{132}Te

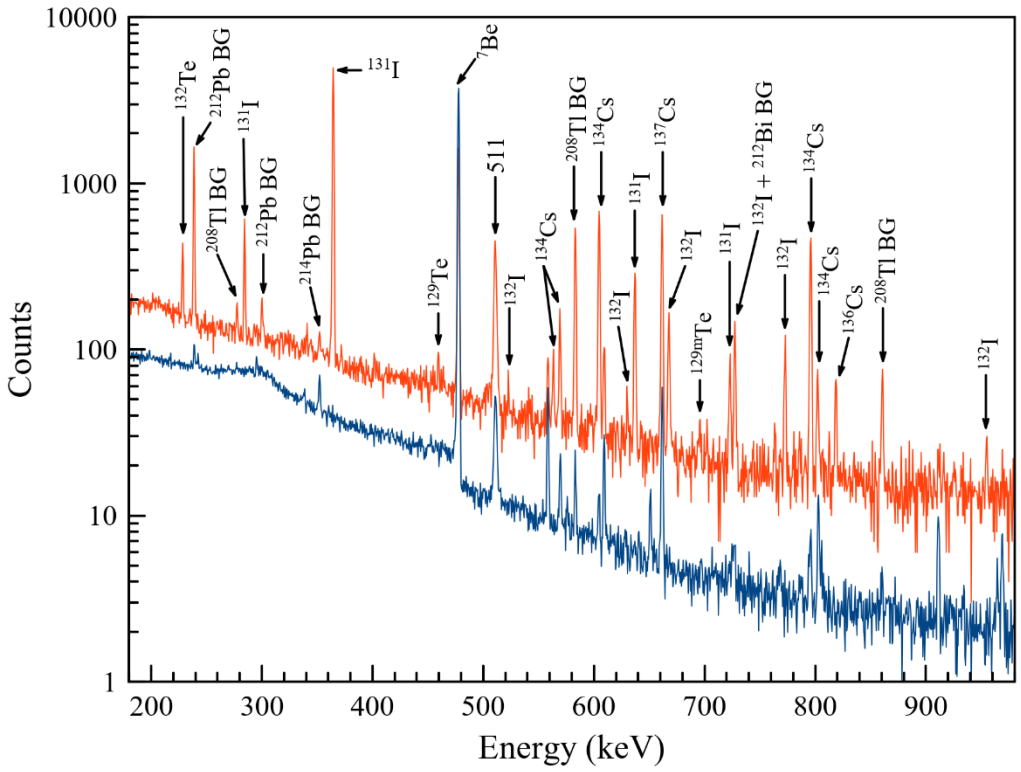

Figure 3. Gamma-ray spectra of the 29 March 2011 TRAC air filter sample showing the peak observed fission product activity (top) compared to the last measurement taken several months later (9 June 2011) (bottom).

(along with its daughter product ^{132}I) and ^{134,136,137}Cs are observed, along with cosmogenically produced ^{7}Be and other radionuclides found in the natural background (e.g., ^{208}Tl, ^{212,214}Pb, and ^{212}Bi). The ^{131}I, ^{134,136,137}Cs, ^{132}Te, and ^{132}I activity concentrations determined from the single-day TRAC sample measurements are given in **Table 1**. The ^{129}Te and ^{129m}Te activity concentrations are not included in tabulated data as the gamma-ray peaks corresponding to these radionuclides were close to the detection limit and quantification uncertain. A correction for cascade summing effects for ^{134}Cs was made using a standard of known activity. The ^{136}Cs and ^{132}I TRAC data reported is uncorrected for cascade summing effects. No summing correction was necessary for ^{131}I, ^{132}Te, or ^{137}Cs.

Table 1. ^{131}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{132}Te and ^{132}I activity concentrations measured with the SRNL TRAC sampler. *Note: ^{136}Cs and ^{132}I data is not corrected for cascade summing effects.

Sample Date	Total SCM	Rainfall (cm)	^{131}I (μBq/SCM)	^{134}Cs (μBq/SCM)	^{136}Cs* (μBq/SCM)	^{137}Cs (μBq/SCM)	^{132}Te (μBq/SCM)	^{132}I* (μBq/SCM)
25-Mar-11	3115	0	552(16)(19)	8.1(33)(3)	< 16(2)	13.6(39)(4)	< 51(5)	< 33(5)
29-Mar-11	5008	0	1488(14)(50)	236(4)(8)	24(2)(1)	231(6)(7)	85(5)(4)	69(5)(2)
30-Mar-11	3388	1.15	188(6)(6)	59(2)(2)	8.2(15)(3)	56(3)(2)	15.7(44)(7)	15.2(30)(4)
1-Apr-11	3771	0	2470(20)(83)	213(4)(7)	21(2)(1)	202(6)(6)	50(7)(2)	21(4)(1)
4-Apr-11	3305	0	233(12)(8)	26(4)(1)	< 17(3)	21.9(50)(6)	< 42(5)	< 31(5)
5-Apr-11	8863	2.77	259(3)(9)	12.2(5)(4)	1.04(41)(3)	12.3(8)(4)	< 8.1(6)	< 3.8(5)
6-Apr-11	16761	0	893(4)(30)	111(1)(4)	6.3(3)(3)	106(1)(3)	6.4(22)(3)	3.7(8)(2)
7-Apr-11	11764	0.15	545(5)(18)	80(1)(3)	4.3(4)(2)	75(1)(2)	< 54(3)	< 19(2)
28-Apr-11	13048	1.30	2.1(6)(1)	0.74(21)(3)	< 1.2(2)	0.77(32)(2)	< 5.1(4)	< 2.1(3)

The total volume of air (in SCM) processed by the filters for each measurement is also given for reference. Except where otherwise noted, uncertainties in measured activity concentrations and ratios from gamma-ray measurements are given according to the convention where the first number in parenthesis represents the statistical uncertainty and the second number the systematic uncertainty; uncertainty magnitudes apply to the last significant digit (0s in whole numbers are

significant) and represent ± 1σ values. The mean $^{134}Cs/^{137}Cs$ activity ratio observed (with individual measurements decay corrected to the time of the Fukushima earthquake (March 11, 2011 14:46 Fukushima time) and weighted based on counting statistics) was 1.069(8)(43). Rainfall amounts given are those measured at the Central Climatology tower (33.2453° N, 81.74° W) at the SRS by the SRNL Atmospheric Technologies Group (Parker and Addis, 1994; Atmospheric Technologies Group, 2022). This tower was located approximately 13 km away from the area where the TRAC data was acquired. The rain gauge is positioned 7 ft (2.1 m) above the ground, measures rainfall in 0.21 mm increments, and records data every 15 minutes. The observed radionuclides and their relative activity concentrations as well as the $^{134}Cs/^{137}Cs$ activity ratio measured agrees well with published data on fission products observed in North America associated with the Fukushima incident (MacMullin et al., 2012; Wetherbee et al., 2012; Thakur et al., 2013; Satou et al., 2018).

The TRAC data was directly compared to the airborne fission product measurements of MacMullin et al. taken at the University of North Carolina at Chapel Hill during a measurement period overlapping the measurements taken at SRNL (MacMullin et al., 2012). The measured activity concentrations at both locations for ^{131}I and ^{137}Cs are shown in **Fig. 4** and **Fig. 5**, respectively. In addition, for ^{131}I, meteorological modeling predictions for the day-to-day radionuclide activity concentrations at SRNL and Chapel Hill, NC are given. Error bars given in graphs represent statistical errors only. Modeling was performed with respect to unit releases and the model data scaled by the maximum activity concentration observed for ^{131}I at Chapel Hill. The simulated maximum was reached on 25 March 2011 at SRNL and on 27 March 2011 at Chapel Hill. The simplified GEM was able to indicate the timing of the plume's appearance in the Southeastern United States to within a few days. Based on the assumed scaling factor, the

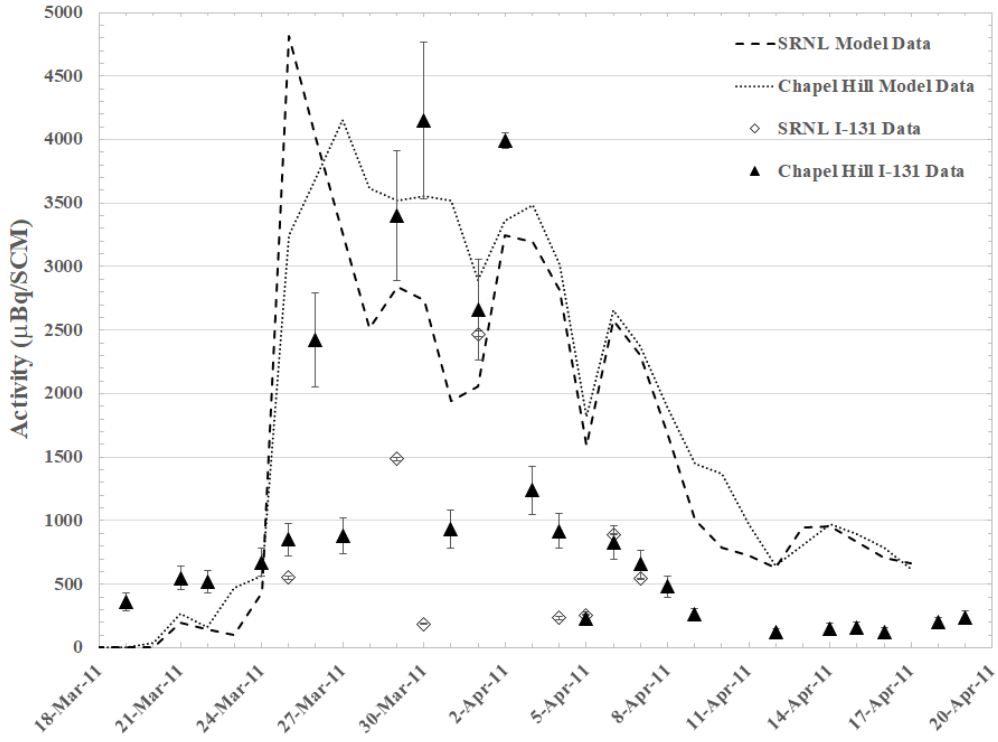

Figure 4. ^{131}I activity concentrations measured at SRNL using the TRAC system and at Chapel Hill, NC compared against model predictions. Model data is normalized to the peak activity observed at Chapel Hill.

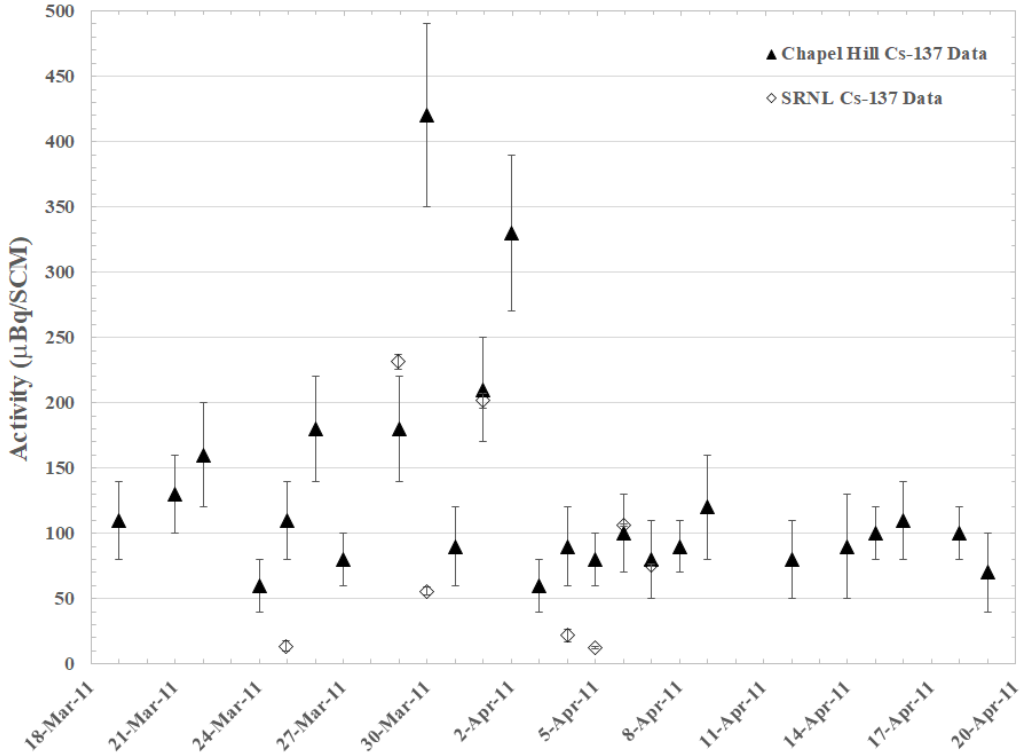

Figure 5. ^{137}Cs activity concentrations measured at SRNL using the TRAC system and at Chapel Hill, NC.

simulations overpredicted measured activity concentrations for SRNL and Chapel Hill on most

days, likely due in part to environmental losses from scavenging processes, but correctly predicted the higher average measured values at Chapel Hill. A trend of elevated activity concentrations from 25 March through the first week of April is observed in both the measured and simulated data. Predicted activity concentrations are very similar for both locations due to the coarse nature of the modeling (SRNL and Chapel Hill, NC are separated by 370 km (230 mi), while grid points are approximately 100 km apart) and the long distances over which the plume spread, allowing for uniform mixing prior to arrival. During the period of maximum measured activity concentrations at SRNL, the SRS experienced several days with significant rainfall. On 30 March 2011, a day on which a large discrepancy between the SRNL and Chapel Hill data was observed, 1.15 cm of rain fell at the SRS (between 9 and 11 am EDT) while 2.44 cm of rain fell at Chapel Hill, NC. There was also heavy (2.77 cm total) rainfall at the SRS in the early morning hours (3:45 am – 6:45 am EDT) of 5 April 2011 with light rain (0.43 cm) at Chapel Hill on the same day. A similar deviation was observed between SRS and Chapel Hill activity concentrations for ^{137}Cs on this date, but measured ^{131}I activity concentrations were identical to within statistical uncertainties. It is not clear based on the limited data available what effect (and its corresponding magnitude) local weather had on relative differences between the activity concentrations of ^{131}I and ^{137}Cs at the two measurement locations.

3.2. Electrostatic Precipitators

The first electrostatic precipitator sample was collected from 23 March through 6 April 2011 for a total of 13.9 days and nominally sampled 199600 SCM of air. As expected, the highest fission product activities and greatest number of fission radionuclides were observed in the sample from this run. None of the shorter-lived radionuclides with half-lives on the order of days (^{129m}Te, ^{132}Te, ^{131}I, ^{132}I, and ^{136}Cs) were observed past the second measurement that was

conducted between 6 April and 19 April 2011 (13.0 days run time and 187070 SCM of air sampled). Subsequent to the fourth measurement (19 May through 21 June 2011, 476230 SCM), ^{134}Cs ceased to be observed and ^{137}Cs activity concentrations approached background levels. The measured activity concentrations for ^{137}Cs, ^{7}Be, and ^{22}Na over the 20-run period spanning from 23 March 2011 to 16 July 2014 is given in **Table 2**. The measured activity concentrations for ^{131}I, ^{134,136}Cs, and ^{129m}Te from 23 March 2011 to 21 Jun 2011 are given in **Table 3**; these radionuclides were undetectable for subsequent measurements. The cascade summing correction

Table 2. ^{137}Cs, ^{7}Be, and ^{22}Na activity concentrations measured with high-volume electrostatic precipitators from 23 March 2011 to 16 July 2014. Note: Due to high count rates, statistical errors were negligible for most ^{7}Be measurements for the stated precision.

Sample Start	Sample Stop	Median Date	Total SCM	^{137}Cs (μBq/SCM)	^{7}Be (mBq/SCM)	^{22}Na (μBq/SCM)
23-Mar-11	6-Apr-11	30-Mar-11	199600	48.6(2)(57)	2.79(0)(17)	0.55(3)(7)
6-Apr-11	19-Apr-11	13-Apr-11	187070	20.2(1)(24)	5.27(1)(32)	1.19(5)(14)
19-Apr-11	19-May-11	4-May-11	430900	3.51(4)(41)	4.15(1)(25)	1.00(3)(12)
19-May-11	21-Jun-11	4-Jun-11	476230	1.17(3)(14)	8.52(1)(51)	1.35(4)(16)
21-Jun-11	21-Jul-11	6-Jul-11	430460	0.179(13)(21)	0.84(0)(5)	0.29(2)(4)
21-Jul-11	22-Aug-11	6-Aug-11	463510	0.138(6)(16)	2.90(0)(17)	0.51(1)(6)
3-Oct-11	14-Nov-11	24-Oct-11	606310	0.089(4)(10)	4.29(0)(26)	0.41(1)(5)
14-Nov-11	28-Dec-11	6-Dec-11	633360	0.107(5)(12)	3.36(0)(20)	0.40(1)(5)
28-Dec-11	6-Feb-12	17-Jan-12	561890	0.104(5)(12)	1.73(0)(10)	0.22(1)(3)
6-Feb-12	28-Mar-12	3-Mar-12	734050	0.137(6)(16)	2.51(1)(15)	0.67(2)(8)
28-Mar-12	30-Apr-12	14-Apr-12	481000	0.116(7)(14)	2.15(1)(13)	0.85(2)(10)
17-Apr-12	6-Jun-12	12-May-12	720170	0.070(3)(8)	1.45(0)(9)	0.65(1)(8)
6-Jun-12	18-Jul-12	27-Jun-12	601900	0.107(6)(12)	1.42(0)(9)	0.56(2)(7)
4-Sep-12	30-Nov-12	17-Oct-12	1252150	0.097(8)(11)	2.84(1)(17)	0.32(2)(4)
30-Nov-12	7-Mar-13	18-Jan-13	1399200	0.110(4)(13)	1.92(1)(12)	0.28(1)(3)
7-Mar-13	10-Apr-13	24-Mar-13	487900	0.113(6)(13)	1.46(1)(9)	0.37(2)(5)
10-Apr-13	24-May-13	2-May-13	635850	0.093(8)(11)	1.49(1)(9)	0.66(3)(8)
30-May-13	15-Aug-13	8-Jul-13	1108800	0.081(5)(9)	1.03(0)(6)	0.33(1)(4)
15-Aug-13	23-Oct-13	19-Sep-13	990300	0.077(4)(9)	2.32(0)(14)	0.28(1)(3)
1-Apr-14	16-Jul-14	24-May-14	1525800	0.078(5)(9)	1.30(1)(8)	0.60(2)(7)

Table 3. ^{131}I, ^{134}Cs, ^{136}Cs, and ^{129m}Te activity concentrations measured with high-volume electrostatic precipitators from 23 Mar 2011 to 21 Jun 2011.

Sample Start	Sample Stop	Median Date	Total SCM	^{131}I (μBq/SCM)	^{134}Cs (μBq/SCM)	^{136}Cs (μBq/SCM)	^{129m}Te (μBq/SCM)
23-Mar-11	6-Apr-11	30-Mar-11	199600	352(1)(41)	49.4(4)(65)	5.00(10)(69)	33(2)(8)
6-Apr-11	19-Apr-11	13-Apr-11	187070	26(1)(3)	19.8(3)(26)	1.05(7)(14)	5.8(9)(13)
19-Apr-11	19-May-11	4-May-11	430900	< 37(2)	3.4(1)(4)	< 1.4(2)	< 4.3(5)
19-May-11	21-Jun-11	4-Jun-11	476230	< 27(1)	0.40(1)(5)	< 1.0(1)	< 4.0(5)

for ^{134}Cs was made using a standard of known activity. Summing corrections for ^{136}Cs were made using a benchmarked MCNP6 (Goorley et al., 2012) simulation. The correction for summing of the 511 keV gamma-ray from position annihilation with the 1274.5 keV gamma-ray emitted by ^{22}Na was estimated based on values measured for detectors with similar response. No summing correction was necessary for ^{7}Be, ^{131}I, ^{129m}Te, or ^{137}Cs. The ^{137}Cs activity concentration is plotted in **Fig. 6**, with the measured ambient ^{22}Na activity concentration (from cosmic ray spallation), which is on the same order, also displayed for comparison. As with the TRAC data plots, error bars shown represent statistical errors only. The uncertainties in activity concentrations given in all graphs (TRAC and electrostatic precipitator) represent one standard deviation from counting statistics. Statistical and systematic errors are given in tabulated data. The dominant sources of systematic error in the TRAC and electrostatic precipitator measurements are given in **Table 4**.

3.3. Plutonium Activity

The total measured plutonium content of the samples, after separation and purification, ranged from 3 to 56 femtograms per sample ($^{239+240}$Pu), with the exception of the 19 May to 21 June 2011 collection which contained over 1250 femtograms of plutonium. The atom fraction ratio of ^{240}Pu to ^{239}Pu measured from each of the electrostatic precipitator samples is shown in **Fig. 7**.

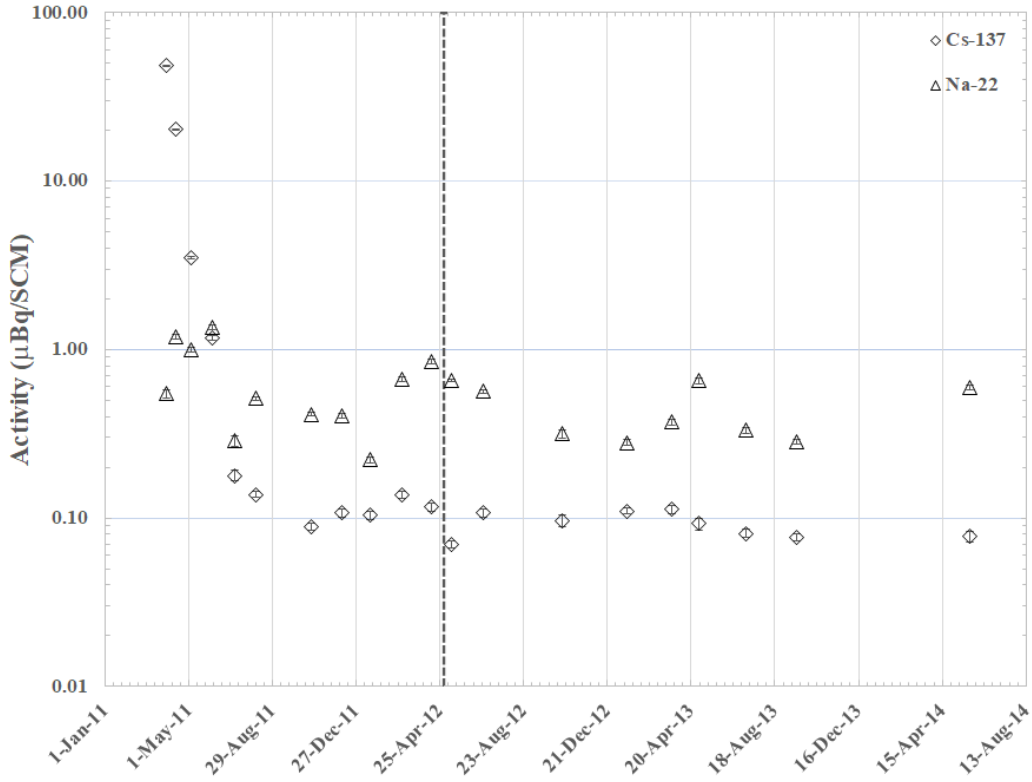

Figure 6. ^{137}Cs and ^{22}Na activity concentrations in air measured at SRNL in the electrostatic precipitator samples. The vertical dashed line indicates the switchover from ground to roof-top level measurements.

Table 4. Major sources of systematic error (± 1σ) in the TRAC and electrostatic precipitator measurements.

Source of Uncertainty	TRAC σ_{sys}(%)	Electrostatic Precipitator σ_{sys}(%)
Detector Efficiency	1.5–3.0	6.0
Branching Ratios	3.4 (^{132}Te)	20 (^{129m}Te)
Cascade Summing Corrections	1.9	2.9–4.5
Collection Efficiency	2.5	–
Air Volume Sampled	–	10

The ground-level electrostatic precipitator measurements showed isotopic compositions of ^{240}Pu that were, on average, lower than global fallout (Kelley et al., 1999); this finding is potentially indicative of ground-level contamination being re-suspended. The measured ratio of ^{240}Pu/^{239}Pu was higher for the roof-top measurements. Two of the samples had ^{240}Pu/^{239}Pu values above 0.3, which likely originated from the processing of high-burnup, legacy materials at the

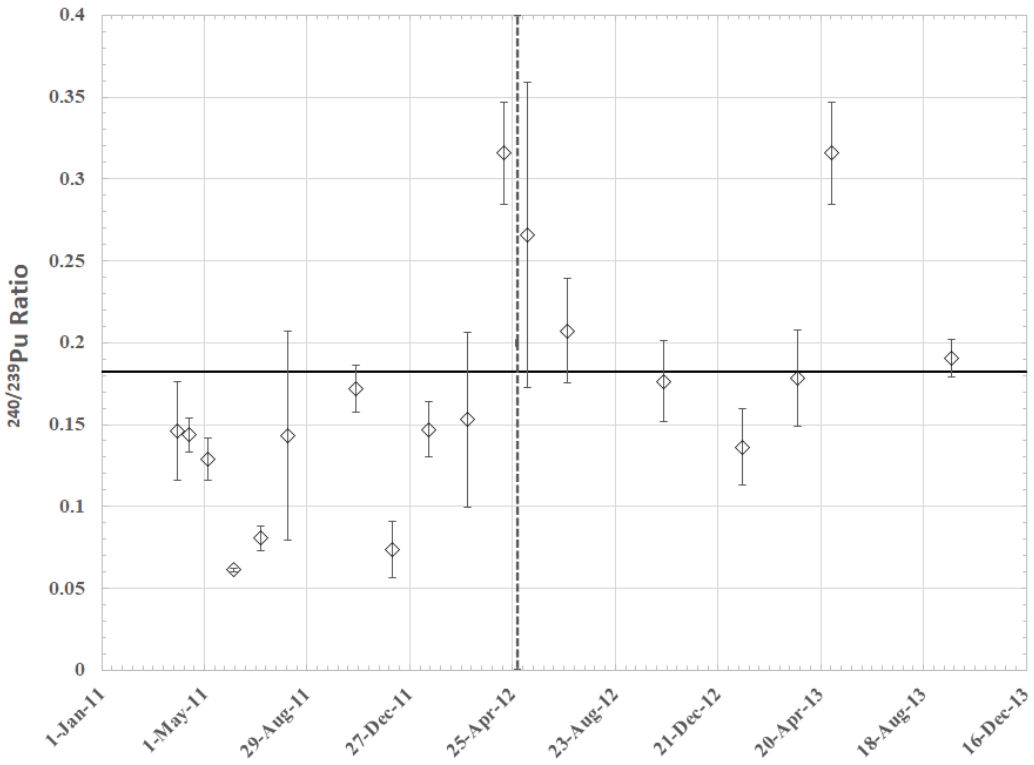

Figure 7. Ratio of ^{240}Pu to ^{239}Pu (by atom fraction) found in SRNL electrostatic precipitator samples. The solid horizontal line indicates the consensus value (Kelley et al., 1999) for global fallout in the northern hemisphere. The vertical dashed line indicates the switchover from ground to roof-top level measurements.

SRS. Measured $^{239+240}$Pu activity concentrations are given in **Table 5**. The ^{241}Pu content of samples was too low to be measured by 3STIMS in most samples.

The largest sample had sufficient activity to measure the ^{238}Pu to $^{239+240}$Pu activity ratio by alpha spectrometry; this ratio was determined to be 0.25 ± 0.02. The ^{240}Pu atom percent of the sample was determined to be < 6%, consistent with weapons-grade material. The ^{241}Pu atom percent was also able to be measured for this sample and was found to be 0.04 ± 0.01, indicating that it was very old material, and thus unrelated to the Fukushima release. The ^{241}Pu content in material from Fukushima was greater than 7 atom percent (Zheng, 2012; Igarashi, 2019). The ^{241}Pu/^{239}Pu ratio measured in this sample and compared to the nominal SRS value of the ^{241}Pu/^{239}Pu ratio for weapons-grade plutonium at time zero (i.e., at the time of its production), gives an age of

Table 5. Start date, total SCM sampled, and $^{239+240}$Pu activity concentrations measured for electrostatic precipitator samples. Uncertainties in the total Pu content represent total uncertainty (± 1σ).

Sample Start	Sample Stop	Median Date	Total SCM	$^{239+240}$Pu (nBq/SCM)
23-Mar-11	6-Apr-11	30-Mar-11	199600	0.35(3)
6-Apr-11	19-Apr-11	13-Apr-11	187070	0.21(1)
19-Apr-11	19-May-11	4-May-11	430900	0.23(1)
19-May-11	21-Jun-11	4-Jun-11	476230	7.04(13)
21-Jun-11	21-Jul-11	6-Jul-11	430460	0.24(2)
21-Jul-11	22-Aug-11	6-Aug-11	463510	0.020(4)
3-Oct-11	14-Nov-11	24-Oct-11	606310	0.27(1)
14-Nov-11	28-Dec-11	6-Dec-11	633360	0.11(1)
28-Dec-11	6-Feb-12	17-Jan-12	561890	0.13(1)
6-Feb-12	28-Mar-12	3-Mar-12	734050	0.07(1)
28-Mar-12	30-Apr-12	14-Apr-12	481000	0.44(4)
17-Apr-12	6-Jun-12	12-May-12	720170	0.06(1)
6-Jun-12	18-Jul-12	27-Jun-12	601900	0.22(2)
4-Sep-12	30-Nov-12	17-Oct-12	1252150	0.07(1)
30-Nov-12	7-Mar-13	18-Jan-13	1399200	0.06(1)
7-Mar-13	10-Apr-13	24-Mar-13	487900	0.08(1)
10-Apr-13	24-May-13	2-May-13	635850	0.16(1)
30-May-13	15-Aug-13	8-Jul-13	1108800	< 0.01
15-Aug-13	23-Oct-13	19-Sep-13	990300	0.10(1)

approximately 50 years for this sample (**Fig. 8**). The nominal isotopic atom percent for this sample was determined to be: 0.105% ^{238}Pu, 94.079% ^{239}Pu, 5.774% ^{240}Pu, and 0.042% ^{241}Pu.

The value of the ^{241}Pu/^{239}Pu ratio was compared with several National Institutes of Standards and Technology (NIST) reference materials of known provenance, including Rocky Flats Soil (SRM 4353a) and Columbia River Sediment (SRM 4350B). The Rocky Flats Soil contained weapons-grade plutonium released in a fire at a processing line at the former Rocky Flats Plant in Colorado, while the Columbia River Sediment contains plutonium released at the Hanford site in Washington state. While the Rocky Flats ratio agreed very well with that measured from the

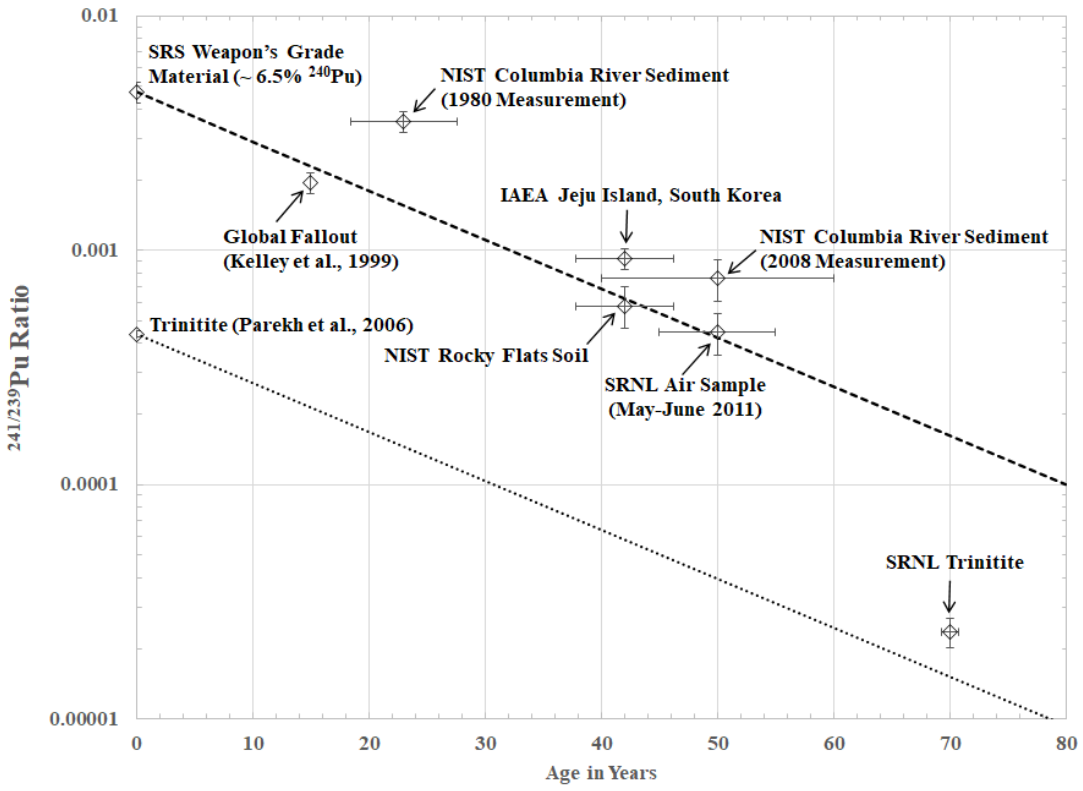

Figure 8. Ratio of ^{241}Pu to ^{239}Pu (by atom fraction) for the SRNL air sample collected from 19 May to 21 June 2011 compared with other global measurements.

SRNL sample, the Columbia River value was somewhat higher. The anomaly in the Columbia River sample has been attributed to an input of fresh ^{241}Pu from fallout from above-ground nuclear testing by the People's Republic of China between 1967 to 1976 (Beasley, et al., 1991). A sample from Jeju Island, South Korea, provided by the International Atomic Energy Agency (IAEA) as a Worldwide Proficiency Sample, IAEA-TEL-2014-03, also shows evidence of Chinese thermonuclear test fallout. As an additional check, and for comparison with a plutonium sample having a different isotopic composition, the ^{241}Pu/^{239}Pu ratio from a sample of Trinitite was also analyzed. The ^{241}Pu content was independently measured by gamma-ray spectrometry of the ingrowth of the ^{241}Am daughter while the ^{239}Pu content was measured by alpha spectrometry. The ^{241}Pu/^{239}Pu ratio of the SRNL Trinitite sample, measured from mass

spectrometric measurements by 3STIMS, agrees quite well with the measurement of Parekh et al., 2006.

4. Conclusions

Fission products released subsequent to the Fukushima Daiichi nuclear accident and observed in the TRAC vehicle data, in order of decreasing activity concentration, included: ^{131}I, ^{134}Cs, ^{137}Cs, ^{129}Te, ^{132}Te (along with its daughter ^{132}I), and ^{136}Cs; this data covered the period from 2 to 7 weeks after the accident. The highest activity concentration of ^{131}I (2470(20)(83) μBq/SCM) was observed on 1 April 2011, while the highest activity concentrations of all other fission products were observed on 29 March 2011. The high-volume, long-collection-time electrostatic precipitator measurements allowed for high-precision continuous monitoring of Fukushima-related and natural radionuclides for several additional years following the accident. Based on this data (see **Table 3**), the level of ^{137}Cs (the longest-lived isotope of the primary radionuclides released) in ground-level air appears to return to the regional background level approximately three months after the accident. The sensitivity of the TRAC and electrostatic precipitator systems was aided greatly by the use of SRNL's ultra-low-background underground counting facility (UCF). The measurements conducted as part of this work provide a valuable additional source of high-quality data for Fukushima radionuclide activity concentrations observed in the Southeast region of the United States, complementing the levels previous measured at Chapel Hill, North Carolina. This data can be used to help refine models for radionuclide transport from planned and unplanned future releases and help set bounds on expected variability in regional activity concentrations measured subsequent to a distant release. All measured activity concentrations for both sampling systems represent particulate-attached species only. Total airborne radionuclide concentrations are expected to be higher for radionuclides that also exist in

a gaseous state (e.g., ^{131}I in the chemical form of I_2). Electrostatic collection significantly simplified radiochemical processing of plutonium samples prior to quantification and increased the sensitivity of low-level detection, particularly for TIMS analysis. The TIMS analysis definitively showed that the largest plutonium sample measured was low-^{240}Pu legacy material, most likely from prior SRS production of weapons-grade material, and not related to plutonium released during the Fukushima event. The remaining plutonium sample amounts and compositions were consistent with a combination of environmental background from global fallout and the ongoing processing of legacy fuel materials at the SRS.

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