Journal of Environmental Management 115 (2013) 217–226

Contents lists available at SciVerse ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Radionuclide activity concentrations in forest surface fuels at the Savannah River Site

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ARTICLE INFO

Article history:
Received 21 October 2011
Received in revised form
19 September 2012
Accepted 8 October 2012
Available online

Keywords:
Litter
Duff
Surface fuels
Radionuclides
Kriging

ABSTRACT

Background/objective: A study was undertaken at the United States Department of Energy's Savannah River Site (SRS), Aiken, South Carolina to investigate radionuclide activity concentrations in litter and duff from select areas at SRS. Litter (i.e. vegetative debris) and duff (i.e. highly decomposed vegetative debris) can often be the major fuels consumed during prescribed burns and have potential to release radiological contaminants into the environment.

Methods: Repeated samples from 97 locations were collected systematically across SRS and analyzed for radionuclide activity. Radionuclide activity concentrations found in litter and duff were compared. As spatial trends were of interest, spatial distributions of radionuclide activity concentrations found in litter and duff and spatial dependency amongst the data were explored.

Results: ^{7}Be, ^{40}K, and ^{137}Cs showed statistically significant proportional differences between litter and duff samples. Duff sample concentrations for ^{137}Cs ($p < 0.0001$) and ^{40}K ($p = 0.0015$) were statistically higher compared to litter samples. ^{7}Be activity concentrations were statistically higher in litter as compared to duff ($p < 0.0001$). For ^{40}K litter and duff samples, spatial correlation tests were not significant at $p = 0.05$ and the maps did not indicate any apparent high concentrations centered near possible radionuclide sources (i.e. SRS facilities). For ^{7}Be litter samples, significant spatial correlation was calculated ($p = 0.0085$). No spatial correlation was evident in the ^{7}Be duff samples ($p = 1.0000$) probably due to small sample size ($n = 7$). ^{137}Cs litter and duff samples showed significant spatial correlations ($p < 0.0001$ and $p < 0.0001$, respectively).

Conclusions: To date, few studies characterize radionuclide activity concentrations in litter and duff, and to our knowledge none present spatial analysis. Key findings show that across SRS, ^{137}Cs is the primary radionuclide of concern, with the highest number of samples reported above MDC in litter (51.4%) and duff samples (83.2%). However, ^{137}Cs litter and duff spatial trends in the maps generated from the kriging parameters do not appear to directly link the areas with higher activity concentrations with SRS facilities. The results found herein provide valuable baseline monitoring data for future studies of forest surface fuels and can be used to evaluate changes in radioactivity in surface fuels in the southeast region of the U.S.

1. Introduction

Previous studies indicate that radionuclides deposited years ago and incorporated into uncultivated soils, floodplains, and surface and ground water can be transported via biological and physical processes into forest floor and woodland vegetation—from root to aboveground tissue—growing on contaminated areas (Adriano and Pinder, 1977; Gürer and Georgopoulos, 2001; McLendon et al., 1976; Pinder et al., 1984; Whicker et al., 1999; Yoschenko et al., 2006a,b; Zhu and Smolders, 2000). Contaminants can incorporate into needles, bark, and leaves that fall and become litter (freshly fallen needles/leaves) (Adriano et al., 1981). Upon further decomposition, this material is termed duff. Both litter and duff can act as highly absorptive mulch, thereby accumulating contaminants (Adriano et al., 1981).

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http://dx.doi.org/10.1016/j.jenvman.2012.10.058

Several global studies investigated the release of radionuclides during forest fires in areas contaminated by nuclear material, including areas contaminated by the Chernobyl Nuclear Power Plant accident (Kashparov et al., 2000; Pazukhin et al., 2004; Yoschenko et al., 2006a,b) and areas near the Los Alamos National Laboratory in the May 2000 Cerro Grande Fire (Johansen et al., 2003; Volkerding, 2004). The radionuclides of greatest concern following Chernobyl were ^{137}Cs, ^{90}Sr, and ^{238,239,240}Pu; however, results showed wildland fires contribute negligibly to the redistribution of radioactivity compared to the whole zone contamination at Chernobyl (Yoschenko et al., 2006a). Radiological emissions released from recent wildfires at Los Alamos, NM showed smoke samples containing naturally occurring elements—which are decay products of radon (^{214}Bi and ^{214}Pb) in the atmosphere (Jacobson, 2001; Whicker et al., 2006).

Due to increased surface area and persistent foliage throughout the year for pine species, airborne particulates are intercepted on the needles (Cooplestone et al., 1999). When the needles fall due to senescence, the litter and duff layers accumulate radioactive materials which have potential to become airborne again during fires (Malilay, 1998). The biogeochemical cycle of each element as well as the generation and decay processes make an assessment of potential exposure complex. Radionuclides can also be taken up from soil by trees and recycled through foliage drop. ^{137}Cs, a common and widely distributed product of nuclear testing and processing, is easily taken up by vegetation as an analog of potassium (Simonoff et al., 2007) and can be volatilized or re-suspended in fires, concentrated in ash (Adriano et al., 1981; Amiro et al., 1996), and redistributed by run-off (Johansen et al., 2003). Varying amounts of ^{137}Cs are found in the environment from nuclear weapons tests that occurred decades ago and from nuclear reactor accidents (CDC, 2004; Peterson et al., 2007). With a half-life of 30.2 years and due to its chemical nature, ^{137}Cs is one of the radioisotopes of major concern at Chernobyl and other nuclear processing facilities like the Savannah River Site (SRS). Naturally occurring radionuclides can also be released into the environment through man-made processes such as mining and burning of fossil fuels (i.e. coal). Coal contains natural radionuclides such as ^{40}K, ^{238}U, and ^{232}Th (Cooper et al., 2003; McBride et al., 1978). At SRS, nuclear facility releases have resulted in the deposition of radionuclides into surface soils (Ellickson et al., 2002). Prescribed burns are conducted routinely at SRS to primarily sustain and improve habitat for the endangered Red-Cockaded Woodpecker (*Piciodes borealis*) and to reduce hazardous fuel accumulation and associated wildfire potential (Kilgo and Blake, 2005). SRS' prescribed burn program pursues a goal of burning approximately 9300 ha per year. Previous studies at SRS determined that total forest floor litter and duff (partially decomposed litter) often contributes to a majority of the fuel consumed during prescribe burns (Goodrick et al., 2010).

The primary objective of this study was to characterize and quantify the radionuclide constituents in litter and duff samples from randomly selected areas within SRS. To date, few studies have characterized radionuclide activity concentrations in litter and duff material. Two previous studies showed elevated concentrations of ^{137}Cs near reprocessing plants in both soil and litter samples, indicating long-term impact of the reprocessing operations on SRS (Adriano and Pinder, 1977; Adriano et al., 1981). A second objective was to understand specific spatial trends of radionuclide activity concentrations and where high activity concentrations are located across SRS. Such data are needed to provide baseline measurements of spatial distributions of radionuclide contaminants found in litter and duff at SRS. Naturally occurring radionuclide activity concentrations in litter and duff are expected to lack spatial correlation within SRS, with no apparent high concentrations near nuclear reactors or major production facilities; while it is hypothesized that anthropogenic radionuclide activity concentrations will be slightly elevated around nuclear reactors and production facilities.

2. Methods

2.1. Study location

The United States Department of Energy (DOE) established SRS for production of special nuclear materials. Construction started at SRS in 1951 with some facilities becoming operational in 1952 (Garten et al., 2000). From 1952 to 1988, SRS' mission was dedicated to the production of nuclear materials, primarily tritium and ^{239}Pu, used in fabrication of nuclear weapons (Cummins, 1994; Dai et al., 2002). Production of nuclear materials discontinued in 1988 and cleanup of contaminated ground water, surface water, sediment, sludge, solid waste, and soil remains in progress (USEPA, 2010).

SRS is also a National Environmental Research Park and is located in the upper coastal plain of South Carolina near Aiken, South Carolina. Approximately 12% of the SRS has been used or is currently used for nuclear processing purposes (ATSDR, 2007). The majority of the remaining portion of the SRS is forested and managed (i.e. prescribed fires) for a variety of operational objectives (ATSDR, 2007; USFS-SR, 2005). At approximately 800 square kilometers (Garten et al., 2000), the forested area of SRS is primarily made up of about 31% hardwood or mixed pine hardwood and 69% pine (Kilgo and Blake, 2005).

2.2. Sample collection

To characterize radionuclide activity concentrations in forest floor samples at SRS, litter (n = 333) and duff (n = 238) samples were collected and analyzed for dry mass and radionuclide activity. Forest floor samples were collected at 97 randomly selected Forest Inventory (FI) sample sites at SRS during the late winter and early spring of 2004 in four major forest types (loblolly/slash pine, longleaf pine, mixed pine and hardwood, and upland hardwood) (Ottmar et al., 2007, Fig. 1). Within each sample site, 4 sub-samples were collected to establish litter and duff loadings across a range of forest conditions.

For each FI sample site, a subplot was designated and marked as the plot center and then located using a combination of global positioning systems (GPS) coordinates and bearing tree information. Four sampling locations were established 10 m (33 feet) from the plot center at each of the four cardinal directions (Fig. 2). If the sampling point proved unrepresentative of the surrounding forest floor, 3 additional meters (10 feet) were added to the original end point until a suitable sample point was located (Ottmar et al., 2007). A 30.5 cm (12 inch) beveled steel square was used to systematically collect and quantify the litter and duff layers. The left corner and left edge of the square were aligned parallel with the cardinal directions and placed on top of the forest floor.

Twelve nails were positioned in a grid pattern within the square, each tapped downwards until the nail was flush with the top of the litter layer. The litter was carefully removed and placed within a labeled bag (FI plot identification, sample type, sub-sample direction, and date). The nails were again tapped down flush with the top of the duff layer. The same procedures from litter collection were administered for duff layer collection. A thorough description of the study design and materials may be found in the congruent study focused on forest floor bulk density evaluation at SRS (Maier et al., 2004).

After collection, all litter and duff samples were initially oven dried for 48 h to determine dry mass: Litter samples at 70 °C and

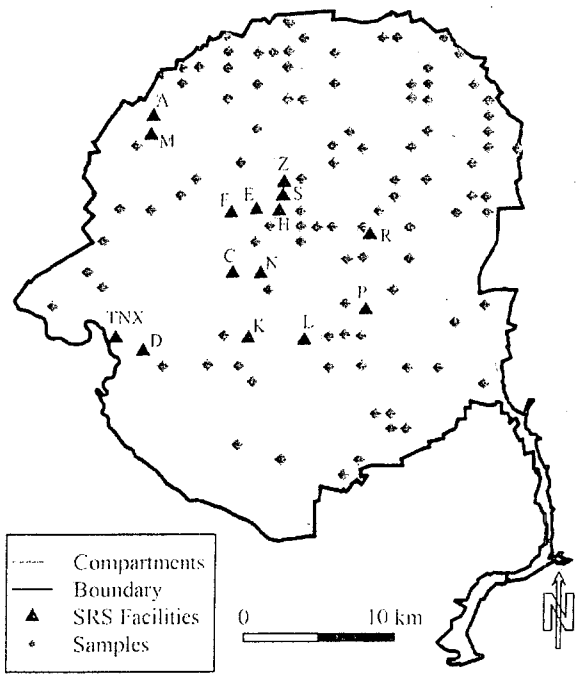

Fig. 1. Litter and duff samples (Forest Inventory [FI] sample sites) across Savannah River Site with plotted SRS facilities. *Compartments* refer to forest management partitions across SRS. *Boundary* refers to the SRS administrative perimeter. *SRS facilities* refer to areas that have released and/or had the potential to release radionuclides (ATSDR, 2007; Carlton, 1999) and thus are identified as major nuclear production facilities and reactors at SRS (ATSDR, 2007), hence:

- C, K, L, P and R—areas with past operating nuclear reactors;
  - P-Area also houses a coal-fired power plant along with coal storage and disposal piles (USEPA, 2011).
- F and H—nuclear processing and separation areas;
- A and M—fuel fabrication areas;
  - A-Area also consists of coal storage and disposal piles (USEPA, 2011).
- E, S and Z—general separation and waste management areas;
- N—storage of construction materials area;
- D—past heavy water reprocessing area, also houses a coal-fired power plant since its construction in 1952 (ATSDR, 2007; USEPA, 2011), and;
- TNX—radionuclide recovery area.

Samples refer to Forest Inventory (FI) sample sites where litter and duff were collected.

duff samples at 100 °C. The samples were then analyzed for radionuclide activity. The samples were analyzed by the SRS Environmental and Bioassay Laboratory (EBL) during the months of June and July of 2004. At the EBL, samples were again placed in a drying oven and allowed to dry overnight or for several hours to assure samples were desiccated before radioactivity measurement.

2.3. Analytical techniques

2.3.1. Gamma spectrometric analysis

Exploratory analysis was performed using gamma spectroscopy for measurement of radionuclide activities suspected to be present in the samples. The vegetation was kept in a Secador Auto-Dessicator to maintain a relative humidity of 25–40% until sample counting and then placed into a calibrated 130G Marenelli beaker or 500 mL low density polyethylene bottle geometry for analysis. The gamma spectroscopy analyses were performed using a Canberra®/Nuclear Data Genie High Purity Germanium Gamma analysis system with either a Canberra® Model GC4019 coaxial germanium detector with a relative efficiency of 40% or a Canberra® Model GC2518 detector with a relative efficiency of 26.7%. Samples were counted for 5000 or 10,000 s and the concentrations of gamma-emitting radioisotopes were determined using the Genie™ applications software. The radionuclide activity concentration results were decay-corrected to the dates when the samples were collected.

The Genie™ software applies the ANSI N42.14 (ANSI, 1999) standard in calculating the quantity of radioactivity in a sample. The ANSI N42.14 forms a solid basis for the routine calibration and use of germanium semiconductor detectors in the measurement of gamma-ray emission rates and, thereby, the activity concentrations of radionuclides in a sample. An efficiency calibration is determined by a least squares fit to a set of efficiency values calculated from empirical measurements from a NIST (National Institute of Standards Technology) traceable standard gamma-standards which scan the energy spectral range. The routine calibration protocol for EBL involves a full energy scale calibration for each high purity germanium detector using a NIST traceable standard. This is performed annually or more frequently as needed such as after installation of new signal process electronics or when the daily quality control check source indicates an out-of-control condition that cannot be corrected. Daily calibration source checks and background checks of the system are performed to ensure the instrument performance has not changed beyond selected limits since the last calibration or background determination.

Radionuclide activity was determined to be significant if the activity was above the minimum detectable concentration (MDC) for each radioisotope within each litter or duff sub-sample. At SRS, the MDCs are calculated at the 95% confidence level using the Curie MDC application (Currie, 1968). MDCs take into account factors such as count time, efficiency of detector, quantity of material analyzed, and background levels measured for a given day. Individual MDC values are not typically reported with the analytical results. The reported "yes" or "no" significance of each result is determined automatically by the analytical software package. Representative MDCs for the soil and vegetation radiological analyses performed at the SRS lab in 2004 are reported in the SRS Environmental Report (2004).

In compliance with EBL protocols, all activity values, significant (above MDC) and non-significant (below MDC) radionuclide activity concentrations were reported for ^{40}K, ^{60}Co, and ^{137}Cs. Only significant activity concentrations were reported for the remaining radionuclide isotopes.

2.4. Statistical procedures

2.4.1. Sign tests

Litter and duff activity concentrations for each isotope were paired by location within each FI sample site. The data were heavily censored with many values falling below their MDC. Therefore, a sign test was used to compare litter and duff activity levels. The sign test is based on the frequency X_1 at which litter activities exceed duff activities and the frequency X_2 at which duff activities exceed litter activities among paired samples. When duff activity exceeds the MDC but the paired litter does not, duff activity level is taken to exceed that of the litter. Conversely, when the litter activity exceeds the MDC but the paired duff does not, litter activity level is taken to exceed that of the duff. When neither duff nor litter activities exceed the MDC, the paired sample provides no information regarding which fuel type had the higher activity, so such paired samples were dropped. Similarly, those samples that were

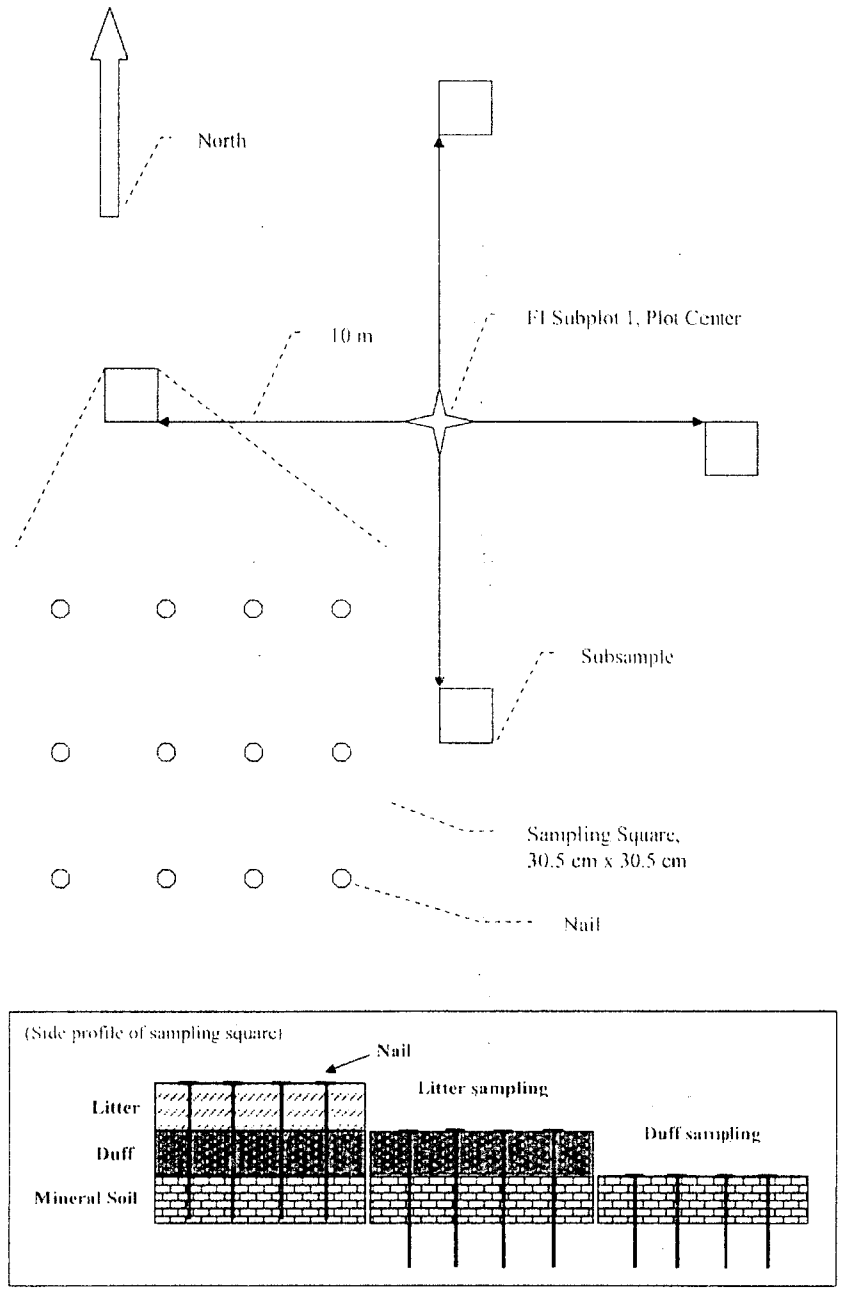

Fig. 2. Typical sample point layout. Adopted and modified from Maier et al. (2004).

not paired were disregarded. Under the null hypothesis that litter and duff activities are equal, the frequencies X_1 and X_2 are expected to be equal. The proportion of paired samples where litter activity exceeds duff activity is

$$\hat{p} = X_1/(X_1 + X_2). \quad (1)$$

Under the null hypothesis, the test statistic

$$z = \frac{\hat{p} - 0.5}{\sqrt{1/4n}} \quad (2)$$

has a standard normal distribution. Comparisons were carried out only for those radionuclides with greater than 20 paired samples. Radionuclides with small sample sizes are addressed in the descriptive statistics only.

2.4.2. Spatial analysis

With no previous spatial data to date, this data allows for the exploration of natural and man-made radionuclide distributions found in litter and duff samples across SRS. The spatial objectives were: i) to identify if radionuclide activity concentrations in litter and duff samples exhibit spatial trends, and; ii) to determine if high radionuclide activity concentrations appear to be located near what

we refer to in this paper as SRS facilities (see Fig. 1), which are identified as major nuclear production facilities and reactors at SRS (ATSDR, 2007). These SRS facilities are defined as areas that have released and/or had the potential to release radionuclides (ATSDR, 2007; Carlton, 1999). As spatial trends were of interest, continuous surface maps for each radionuclide were created using ordinary kriging to explore spatial distributions in litter and duff radionuclide activity concentrations. Spatial correlation tests were also performed to identify if significant spatial dependency existed in the data.

The data used in this study included the spatial coordinates for each field sample (collected with GPS), the locations of SRS facilities, forest management compartments, and the SRS administrative boundary which was provided by the United States Forest Service-Savannah River (USFS-SR). All geographic data used in this study were spatially referenced using the 1927 North American Datum and then projected using the Universal Transverse Mercator (UTM) zone 17N coordinate system.

It is important to note that as individual MDC values were not provided in the lab reported results (except ^{40}K, ^{60}Co, and ^{137}Cs), a soil/sediment generic MDC of 1.06×10^1 Bq/kg (original figure in pCi, converted to Bq using 1 Bq = 27 pCi) (retrieved from SRS Environmental Report, 2004 – Table 2: Representative Minimum Detectable Concentrations for Radiological Analyses) was used for spatial locations that were below the limit of detection for ^{7}Be.

The spatial analysis was conducted in two stages. First, spatial covariance for each data set was modeled using an exponential variogram constructed by restricted maximum likelihood (REML) in the *PROC MIXED* procedure of SAS (Littell et al., 1996) version 9.2 (Cary, N.C.). Spatial dependence among observations was described by fitting the exponential variogram to the data,

$$2\gamma(d) = c_0 + c_1(1 - \exp\{-d/a\}) \quad (3)$$

where γ is the theoretical variogram; d is the distance between data points; c_0 is the nugget effect; c_1 is the partial sill; and α is the range parameter.

The variogram assumes that the strength of spatial dependence depend only on the distance d between a pair of sites. The nugget effect, c_0, describes the small-scale spatial variation, variation at scales shorter than the distances between neighboring sites. The sill, $c_0 + c_1$, is equal to twice the population variance of the data, and the range, α, is the distance at which the difference between the variogram from the sill becomes negligible. Pairs of sites further than distance 3α apart are negligibly correlated.

Second, ordinary kriging (Webster and Oliver, 2001) (based on the previously described spatial models) was used to interpolate radionuclide concentrations across a grid constrained by the point sample locations. Ordinary kriging was performed using the *krigconv* function in the 'geoR' package and final maps were created using the 'map' and 'maptools' packages in R (R Development Core Team, 2010).

3. Results

3.1. Radionuclide activity concentrations

The mean, standard deviation, 95% confidence interval (CI), sample size, number and percent above MDC, and minimum and maximum observed specific activity concentration of all identified radionuclides are reported in Supplementary Tables 1 and 2 for the litter and duff samples, respectively. ^{208}Tl, ^{212}Pb, ^{214}Pb, ^{212}Bi, and ^{214}Bi were detected in a majority of the litter and duff samples (Supplementary Tables 1 and 2). However, these radionuclides are short-lived decay products of radon, an inert gas, found naturally in the environment. Due to their short half-lives and because of the long delay between sampling and analyses, the radionuclide activity concentrations reported are not indicative of what is in the sample and therefore are not included in the further analysis.

Table 1
Sign test table for ^{7}Be, ^{60}Co, ^{40}K, and ^{137}Cs.

Radionuclide	Litter (X_1)[a]	Duff (X_2)[b]	$\hat{p}$[c]	Z statistic[d]	p-Value[e]
^{7}Be	62	2	0.97	13.29	**<0.0001**
^{60}Co	87	114	0.43	−1.90	0.0569
^{40}K	78	123	0.39	−3.17	**0.0015**
^{137}Cs	18	183	0.09	−11.64	**<0.0001**

[a] Frequency at which litter activities exceed duff activities among paired samples.
[b] Frequency at which duff activities exceed litter activities among paired samples.
[c] Proportion of paired samples where litter activity exceeds duff activity.
[d] Test statistic under the null hypothesis.
[e] p-Value < 0.05 is significant.

Additionally, ^{228}Ac, ^{224}Ra, ^{231}Th, ^{234}Th, and ^{234m}Pa, were reported in the original data set, but these radionuclides are short-lived progeny which only exist in secular equilibrium with their parent radionuclide, they do not exist on their own. Therefore, for this analysis herein, ^{228}Ac was reported as ^{228}Ra, ^{224}Ra as ^{228}Th, ^{231}Th as ^{235}U, and ^{234}Th and ^{234m}Pa were reported as ^{238}U. Although, progeny of ^{238}U were detected, they were detected in minimal samples (3 samples in litter and 2 samples in duff), so data for them were not considered for further statistical analysis. However, for completeness and future comparative references, activity concentrations for all radionuclides measured in litter and duff during 2004 are provided in Supplementary Tables 1 and 2, respectively. Thereafter, proportional comparison results are only displayed for the following radionuclides—^{7}Be, ^{60}Co, ^{40}K, and ^{137}Cs (Table 1). Three of the four radionuclides—^{7}Be, ^{40}K, and ^{137}Cs—showed statistically significant proportional differences between litter and duff samples (Table 1).

Litter and duff mean radionuclide activity concentrations ^{7}Be, ^{40}K, and ^{137}Cs are displayed in Fig. 3. ^{137}Cs ($p < 0.0001$) and ^{40}K ($p = 0.0015$), had statistically higher activity concentrations in duff compared to litter samples (Table 1). The mean ^{137}Cs activity concentration in litter was 9.89×10^0 Bq/kg (95%CI: 9.00×10^0, 1.07×10^1 Bq/kg) and 2.36×10^1 Bq/kg (95%CI: 2.16×10^1, 2.56×10^1 Bq/kg) for duff (Fig. 3). The mean ^{40}K activity concentration found in litter and duff were 2.43×10^1 Bq/kg (95% CI: 1.70×10^1, 3.17×10^1 Bq/kg) and 4.22×10^1 Bq/kg (95%CI: 3.26×10^1, 5.22×10^1 Bq/kg), respectively (Fig. 3). ^{7}Be activity concentrations were, however, statistically higher in litter samples verses duff ($p < 0.0001$; Table 1). Mean activity concentration for

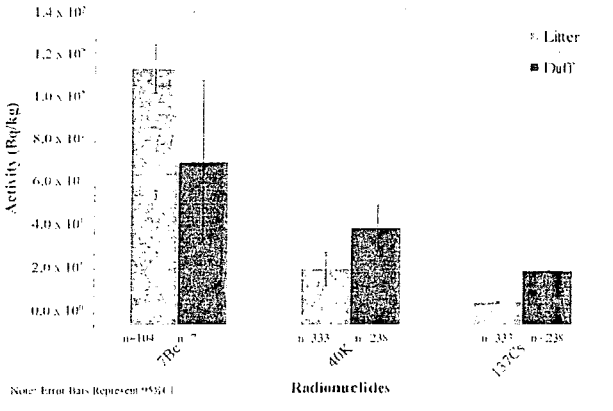

Fig. 3. Litter and duff mean radionuclide activity concentrations for ^{7}Be, ^{40}K, and ^{137}Cs.

^{7}Be in litter was 1.13×10^{2} Bq/kg (95%CI: 1.02×10^{2}, 1.24×10^{2} Bq/kg) and 7.11×10^{1} Bq/kg (95%CI: 3.47×10^{1}, 1.08×10^{2} Bq/kg) in duff (Fig. 3). ^{60}Co did not show significant differences between litter and duff samples ($p = 0.0569$; Table 1).

3.2. Spatial distributions

Kriging parameters and corresponding mean activity concentrations, standard error, degrees of freedom, p-values, and chi-squared values are provided in Table 2. Significant spatial correlation existed in ^{7}Be litter samples ($p = 0.0085$, $n = 104$) and in both ^{137}Cs litter and duff samples ($p < 0.0001$, $n = 333$ and $p < 0.0001$, $n = 238$, respectively) (Table 2). No significant spatial correlation was found in ^{40}K litter or duff samples ($p = 0.6611$, $n = 333$ and $p = 0.9644$, $n = 238$ respectively) or in ^{7}Be duff samples ($p = 1.0000$, $n = 7$) (Table 2). Fig. 4 presents the final continuous surface maps for the litter and duff samples of ^{7}Be, ^{40}K, and ^{137}Cs.

Although ^{60}Co is a concern at DOE sites due to its longer half-life of about 5.3 years and external hazardous gamma rays during its radioactive decay process (Peterson et al., 2007), all litter and duff ^{60}Co activity concentrations were below MDC and could not be included in spatial analyses. SRS soil and vegetative data for ^{60}Co activity concentrations are available for 2004 (SRS Environmental Report, 2004). Given the limited sample size for the remaining radionuclides, kriging models could not be generated.

4. Discussion

This study investigated radionuclide activity concentrations and spatial distributions of radionuclides in litter and duff across SRS. Continuous surface maps are discussed identifying any spatial correlations and are visually assessed to address whether high activity concentrations are relative to potential sources (i.e. SRS facilities). Due to the relative lack of litter and duff data in the literature, much of the referential data presented herein are from soil radionuclide activity concentrations. The radionuclides ^{137}Cs, ^{40}K, and ^{7}Be are discussed because of results and levels found in litter and duff samples across SRS in 2004.

4.1. Anthropogenic radionuclide

4.1.1. Cesium-137

^{137}Cs is the most abundant radiological soil contaminant at SRS (SRS Environmental Report, 2008). Results of a previous study show that the average activity concentration is approximately 1.11×10^{0} Bq/kg (original figure in pCi, converted to Bq using 1 Bq = 27 pCi) in SRS soil samples ($n = 38$) and is consistent with local background levels (Kubilius et al., 2004). The SRS Environmental Report provides soil ^{137}Cs activity concentrations, taken during the Savannah River Swamp Surveys, ranging from non-detectable to 1.85×10^{3} Bq/kg ($n = 39$) (SRS Environmental Report, 2004). Our results showed litter (9.89×10^{0} Bq/kg [95%CI: 9.00×10^{0}, 1.07×10^{1} Bq/kg]) and duff (2.36×10^{1} Bq/kg [95%CI: 2.16×10^{1}, 2.56×10^{1} Bq/kg]) average activity concentrations were both higher than the average reported for the soil in the Kubilius study (2004), but were within range of the Savannah River Swamp Survey soil samples.

Higher ^{137}Cs activity concentrations were measured in the duff samples compared to the litter in the current study (Table 1, Fig. 3). Out of 238 duff samples, 198 samples (83.2% above MDC) contained significant ^{137}Cs activity while 171 out of 333 litter samples (51.4% above MDC) were found to have significant ^{137}Cs activity. A possible explanation of this finding could be attributed to lignin. It has been studied that plants with higher lignin content tend to have higher concentrations of ^{137}Cs; ^{137}Cs can accumulate in lignin upon plant uptake by as much as 90% in hardwood (Andolina and Guillitte, 1990; Rafferty et al., 1997). Though not measured, the duff layer may have had more lignin than the litter layer at SRS. Rafferty et al. (1997) found that ^{137}Cs retention is higher in more humified litter which is decomposed litter or in this case duff (Berg and McClaugherty, 2007; Ottmar et al., 2007). When litter decomposes, it has been shown that lignin content increases in decomposed organic material (Coûteaux et al., 1995).

The activity concentration of ^{137}Cs in surface soil from global fallout events ranges from approximately 3.70×10^{0} Bq/kg to 3.70×10^{1} Bq/kg (Peterson et al., 2007). SRS litter and duff ^{137}Cs average activity concentrations fall within range in soil surfaces from global fallout. Cesium preferentially adheres to soil, particularly sandy and clay soils, and is generally one of the less mobile radioactive elements within the environment (Peterson et al., 2007). The maximum ^{137}Cs activity concentration, 1.12×10^{2} Bq/kg, was found in the duff (more than 3 times the higher end of what is usually found in soils from global fallout events). In an earlier study looking at ^{137}Cs contamination in and around swamp areas at SRS, a range of ^{137}Cs activity concentrations varying from 5.00×10^{0} Bq/kg to 2.40×10^{3} Bq/kg was detected in soil samples (Fledderman et al., 2007) and consequently the maximum activity concentration found in the 2004 duff samples is within range of previous soil measurements collected from SRS.

Table 2
Kriging parameters and corresponding mean activity concentrations, standard error of the mean, degrees of freedom, p-values, and chi-squared values for ^{7}Be, ^{40}K, and ^{137}Cs litter and duff samples.

Parameter	Radionuclide (Litter, Duff)[a]		
	^{7}Be[b]	^{40}K	^{137}Cs
Range (km)	5.92×10^{0}, 5.69×10^{-1}	6.57×10^{1}, 3.33×10^{4}	2.00×10^{0}, 1.63×10^{0}
Nugget (Bq/g)^2	2.84×10^{-3}, 1.65×10^{-4}	4.64×10^{-3}, 5.93×10^{-3}	3.35×10^{-5}, 1.58×10^{-4}
Partial Sill (Bq/g)^2	4.20×10^{-4}, 0	1.73×10^{-4}, 1.84×10^{-2}	3.54×10^{-5}, 1.10×10^{-4}
Mean (Bq/kg)	4.11×10^{1}, 1.24×10^{1}	2.66×10^{1}, 4.26×10^{1}	9.85×10^{0}, 2.39×10^{1}
SE (Bq/kg)	4.93×10^{0}, 8.44×10^{-1}	1.04×10^{1}, 1.36×10^{2}	7.63×10^{-1}, 1.49×10^{0}
Degrees of freedom	2, 1	2, 2	2, 2
X^2	9.53, 0	0.83, 0.07	82.35, 25.11
p-Value	**0.0085**, 1.0000	0.6611, 0.9644	**<0.0001**, **<0.0001**

Note: Exponential variograms were fitted to the data to test whether spatial dependency existed. *Range (km)* represents the distance beyond which there is little or no spatial correlation. *Nugget (Bq/g)*^{2} is the measurement of effect and gives the microscale variation at spatial scales too fine to detect. *Partial Sill (Bq/g)*^{2} is the difference between the nugget and the sill (the overall variability of the data). *SE (Bq/kg)* is the standard error. χ^2 is the chi square test statistic. *p-Value* < 0.05 is significant.

[a] Litter values are represented first while duff values are presented second.

[b] As individual MDC values were not provided in the lab reported results, a soil/sediment generic MDC of 1.06×10^{1} Bq/kg (retrieved from SRS Environmental Report, 2004; original figure in pCi, converted to Bq using 1 Bq = 27 pCi) was used for spatial correlation calculations for all below detect values (in the case of ^{7}Be).

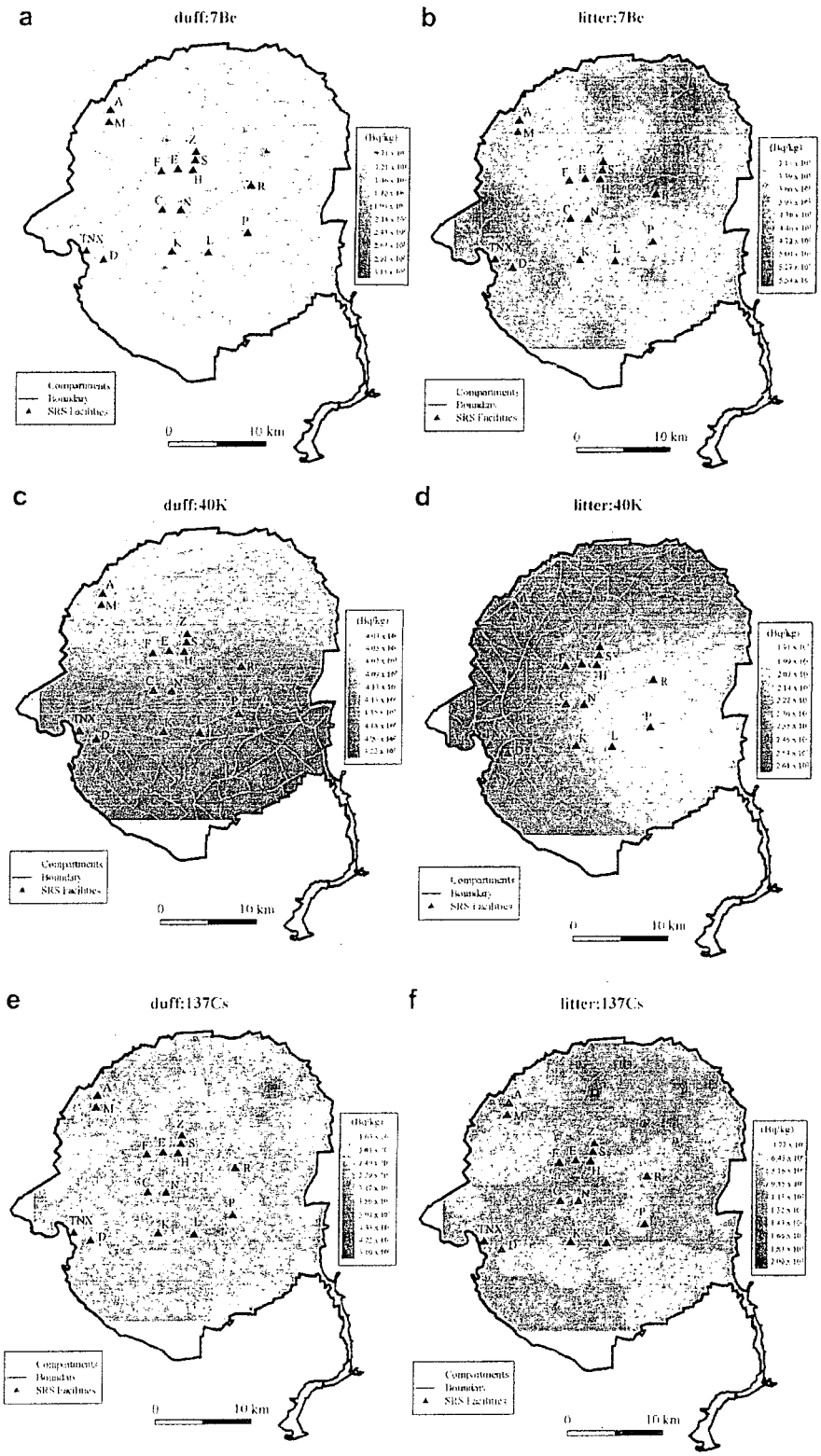

Fig. 4. Continuous surface maps of radionuclides (^{7}Be, ^{40}K, ^{137}Cs) generated using ordinary kriging. Note: Maps are derived based on litter and duff sampling locations (Forest Inventory sample sites [Fig. 1]: ◆) and not *SRS facilities*. The 15 plotted *SRS facilities* (A, C, D, E, F, H, K, L, M, N, P, R, S, TNX, and Z) are defined in Fig. 1. Map contours are displayed using 10 equal intervals across final model predictions. Contour intervals were taken from the minimum and maximum values of the data for each radionuclide (^{7}Be, ^{40}K, and ^{137}Cs). (a) ^{7}Be duff map. No significant spatial correlation exists ($p = 1.000$; $n = 7$) due to small sample size above minimum detectable concentration. (b) ^{7}Be litter map. Spatial correlation is significant ($p = 0.0085$; $n = 104$). (c) ^{40}K duff map. Spatial correlation is insignificant ($p = 0.9644$; $n = 238$). (d) ^{40}K litter map. Spatial correlation is insignificant ($p = 0.6611$; $n = 333$). (e) ^{137}Cs duff map. A significant spatial correlation exists ($p < 0.0001$; $n = 238$). (f) ^{137}Cs litter map. A significant spatial correlation exists ($p < 0.0001$; $n = 333$).

Spatial correlation tests found significant spatial correlations in ^{137}Cs litter and duff samples (Table 2); however, the ^{137}Cs litter and duff spatial trends in the maps generated from the kriging parameters do not appear to directly link the areas with higher activity concentrations with SRS facilities (Fig. 4(e) and (f)). Concentrations found in duff samples showed that higher concentrations appeared near the center, northern, and southerly regions of the SRS with a number of samples with highest activity values being present in the northeastern portion of the map (Fig. 4(e)). Higher ^{137}Cs activity concentrations in litter samples appear near the center and the northeastern regions of the SRS (Fig. 4(f)). Such an occurrence may be due to previous fallout from nuclear weapons testing in past years and possible contamination from SRS sources (Corey et al., 1982). In the past, atmospheric emissions from reactors and other processing stacks (located near the center region of SRS [Fig. 1]) released radionuclides onto surface soils at SRS (Ellickson et al., 2002). Auspiciously, SRS releases have been reduced greatly since the end of the cold war in 1991 and global fallout has decreased greatly on the SRS (as throughout the U.S.) since weapons testing ending in the 1960's (SRS Environmental Report, 2008). Additionally, with time, ^{137}Cs levels are expected to decrease due to radioactive decay. The spatial distributions that were observed in the current study could also be a corollary of environmental factors such as the uptake of ^{137}Cs by plants as it is easily taken up by vegetation as an analog of potassium (Simonoff et al., 2007). Though not measured, percent canopy coverage could play a role. Higher activity concentrations may be due to interceptive function of woodland canopy (Cooplestone et al., 1999).

4.2. Naturally occurring radionuclides

4.2.1. Potassium-40

Although naturally occurring, ^{40}K can also be released into the environment by man-made processes through the mining and burning of coal which naturally contains ^{40}K (Cooper et al., 2003; McBride et al., 1978). D-Area, once a heavy water reprocessing area at SRS, has had an operating coal-burning power plant since its construction in 1952 and is planned to be shut down in 2011 (ATSDR, 2007; USEPA, 2011). Since the early 1950's, there has also been coal storage and disposal piles within SRS facilities such as P-Area (also housing a coal-burning power plant) and A-Area (USEPA, 2011). Such processes may have elevated ^{40}K levels found at SRS.

The natural activity concentrations usually found in soil range from 3.70×10^1 to 1.11×10^3 Bq/kg (HPSUM, 2005). The maximum activity concentration of ^{40}K found at SRS was 5.89×10^2 Bq/kg in duff and falls within the natural activity soil range. SRS available soil data has ^{40}K average activity concentration of 4.52×10^1 Bq/kg (Range: 1.17×10^1, 4.52×10^2 Bq/kg) and vegetative data of 4.70×10^2 Bq/kg (Range: 6.37×10^1, 1.23×10^3 Bq/kg) (SRNS, 2011); average ^{40}K litter and duff activity concentrations are lower than soil and vegetative data and also fall within the range of normal soil global concentrations (HPSUM, 2005; UNSCEAR, 2000). ^{40}K mean activity concentrations were statistically higher in duff compared to litter samples across SRS (Table 1; Fig. 3). This phenomenon is plausible because ^{40}K assimilates into the tissues of living organisms and plants and readily adheres to soil particles, behaving in the environment like other potassium isotopes (Peterson et al., 2007). Duff material more closely resembles soil particle composition—comprised of dense, well-compacted, moderately to highly decomposed organic material—when compared to litter, which is comprised of little to no decomposed material, with its original plant structures still recognizable.

Spatial correlation tests found no significant spatial correlations for ^{40}K litter and duff (Table 2). Lack of spatial correlation in the distribution at SRS was to be expected due to the natural occurrence of ^{40}K in the environment. Based on visual assessment, continuous surface maps generated from the kriging parameters do not appear to directly link the areas with higher activity concentrations with SRS facilities (Fig. 4(c) and (d)). Contours indicate that higher concentrations found in duff samples appear to be localized near the southeastern region of the SRS (Fig. 4(c)). Higher ^{40}K activity concentrations in litter samples appear near the west and northwestern regions of the SRS (Fig. 4(d)).

4.2.2. Beryllium-7

^{7}Be is a naturally occurring radionuclide, produced by cosmic-ray interactions with atmospheric constituents (Matisoff et al., 2002). Our results show average litter and duff ^{7}Be activity concentrations higher than natural activity levels (0.01 Bq/kg; HPSUM, 2005). After its formation in the stratosphere, ^{7}Be adsorbs electrostatically to aerosols and is transported and deposited over exposed surfaces through various physical processes (i.e. wet and dry deposition) (Kaste et al., 2002); hence, ^{7}Be prevailingly accumulates on vegetation canopy and litter in forest areas.

Historical SRS soil data did not have any samples above MDC, while vegetative data shows average ^{7}Be activity concentrations at 1.61×10^2 Bq/kg (Range: 2.64×10^1, 7.04×10^2 Bq/kg) (SRNS, 2011). Average litter and duff ^{7}Be activity concentrations are slightly lower than vegetative values. Higher activity concentrations were often found in litter samples compared to duff (Table 1). This result could be expected due to ^{7}Be fallout from the atmosphere; litter is the first point of contact (Adriano et al., 1981; Cooplestone et al., 1999). Due to its shorter half-life (53.3 days), ^{7}Be may decay prior to the litter decomposing into duff. Further, the different proportions of ^{7}Be versus ^{137}Cs in litter and duff can be explained in consideration of the radioactive decay and of the complex mechanisms of vertical migration and of accumulation.

No significant spatial correlation was observed for ^{7}Be in duff samples (Table 2), probably due to the small number of samples with detections above MDC ($n = 7$). No visual assessment can be concluded for ^{7}Be duff map (Fig. 4(a)) because of the small number of sample size with ^{7}Be detections above MDC. However, the litter map shows a significant spatial correlation (Table 2). Spatial trends in the ^{7}Be litter continuous surface map generated from the kriging parameters do not appear to link areas that have higher activity concentrations with SRS facilities (Fig. 4(b)). ^{7}Be activity concentrations found in litter samples appear to be concentrated in the west, southwest and north, northeastern regions of SRS with a few high values intermittently dispersed within the SRS (Fig. 4(b)). Given that ^{7}Be is naturally occurring, man-made emissions or deposition could not have been responsible for the observed spatial variation. The surface distribution of ^{7}Be in litter prevailingly depends on the spatial distribution of wet precipitations, on the interception of rain by the vegetation canopy and on the patterns and the features of water run-off over the ground for a relatively short period of time before the sampling instant, in view of the half-life of ^{7}Be.

4.3. Limitations

Gross alpha and gross beta measurements in litter and duff samples were not measured and so quantitative levels of radionuclides expected to contribute to gross alpha and beta measurements could not be determined. Used as baseline, gross alpha and beta measurements can help decipher atypical radionuclide activity concentrations found in a select area (Hernández et al., 2005).

The spatial analysis performed was intended to provide a broad overview of radionuclide concentrations across the SRS. Therefore, results are limited to examine trends across the entire SRS (i.e.

global) and not trends that may exist at small-scale (i.e. local) around individual SRS facilities; nevertheless, the litter and duff data can be used to make inferences in this regard.

Furthermore, no control locations were measured to compare litter and duff radiological activity concentrations elsewhere in states near the Atlantic Coastal Plain or other regions of the southeastern United States. Future areas of interest include further exploration in the release, fate, and transport of radionuclides found in concurrent soil, litter, duff, and smoke samples from fire occurrence at SRS. Future research may lead to better trapping technology and more accurate predictive modeling of environmental deposition of radionuclides (Engelmann et al., 2009).

5. Conclusion

To date, few studies characterize radionuclide activity concentrations in forest fuel bed material such as litter and duff, and to our knowledge none present spatial analysis. Our results allow for the exploration of natural and man-made radionuclide distributions found in litter and duff samples particular to the southeast region of the U.S. Moreover, the results found herein provide valuable baseline monitoring data for future studies of forest surface fuels and can be used to evaluate changes in radioactivity in surface fuels in the southeast. Litter and duff monitoring data may be helpful in understanding the forest ecosystem cycling of radioactive contaminants, especially ^{137}Cs.

Key findings show that across SRS, ^{137}Cs, is the primary radionuclide of concern, with the highest number of samples reported above MDC in litter (51.4% above MDC) and duff samples (83.2% above MDC). These data were used at SRS for confirmation of dose assessments performed in support of controlled burns (Lee and Hunter, 2006). Naturally occurring radionuclides ^{40}K and ^{7}Be (except ^{7}Be litter samples) showed no significant spatial correlations across the collection area; whereas anthropogenic ^{137}Cs showed significant spatial correlations in litter and duff samples across SRS. However, the ^{137}Cs litter and duff spatial trends in the maps generated from the kriging parameters do not appear to directly link the areas with higher activity concentrations with SRS facilities.

Acknowledgments

Funding and support was provided by the Department of Energy-Savannah River Operations Office through the U.S. Forest Service Savannah River under Interagency Agreement DE-AI09-00SR22188. Sincere appreciation goes to John Blake, Pete Fledderman, Don Faison, Jay Hutchison, Sherrod Maxwell, Dan Shea, Brian Maier, Clint Wright and crew.

Appendix A. Supplementary material

Supplementary material related to this article can be found at http://dx.doi.org/10.1016/j.jenvman.2012.10.058.

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