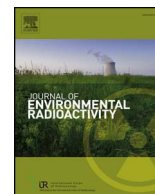




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Radionuclide distribution in soil and undecayed vegetative litter samples in a riparian system at the Savannah River Site, SC

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ABSTRACT

The aim of this study is to comprehensively investigate radionuclide concentrations in surface soil and undecayed vegetative litter along four stream systems (i.e. Fourmile Branch, Lower Three Runs, Pen Branch, and Steel Creek) at the Savannah River Site (SRS), Aiken, South Carolina. Soil and litter samples from systematically spaced 12 pairs (contaminated or uncontaminated) of plots along the four streams were analyzed for 16 distinct radionuclide activities. Lower radionuclide concentrations were observed in soil and litter samples collected along Pen Branch compared to the other 3 streams. The anthropogenic radionuclide with the highest activity was ¹³⁷Cs in soil (10.6–916.9 Bq/kg) and litter (8.0–222.3 Bq/kg), while the naturally occurring radionuclides possessing the highest concentration in the samples were ⁴⁰K (33.5–153.7 Bq/kg and 23.1–56.0 Bq/kg in soil and litter respectively) and ²²⁶Ra (55.6–159.9 Bq/kg and 30.2–101.8 Bq/kg in soil and litter respectively). A significant difference ($p < 0.05$) of radionuclide concentrations between paired-plots across four streams was observed for ²⁴¹Am, ¹³⁷Cs, ²³⁸Pu, ²³⁹Pu, and ²²⁶Ra in both contaminated and non-contaminated samples. ¹³⁷Cs and uranium isotopes had the highest litter-to-soil correlation in contaminated ($\rho = 0.70$) and uncontaminated plots ($\rho = 0.31$ – 0.41), respectively. ⁹⁰Sr was the only radionuclide with higher radioactive concentrations in litter (12.65–37.56 Bq/kg) compared to soil (1.61–4.79 Bq/kg). The result indicates that 1) historical discharges of anthropogenic ¹³⁷Cs was the most important contributor of radiation contamination in the riparian environment at SRS, 2) ⁹⁰Sr was the only radionuclide with higher concentration in litter than in soil, and 3) no apparent pattern in deposition density in soil or litter along downstream was observed for the radionuclides measured in this study.

1. Introduction

Artificially produced radionuclides, such as ²⁴¹Am and ¹³⁷Cs, which are operationally discharged from nuclear facilities into the environment, can be accumulated into forest soils (Schimmack et al., 1994). These could in turn be remobilized into other environmental compartments including ground and surface water, vegetation through plant uptake and the atmosphere through particle resuspension, and volatilization during wildfires (Carvalho et al., 2014; Ehken and Kirchner, 2002; Hejl et al., 2013; Lux et al., 1995; Mamikhin et al., 1997; Xu et al., 2011, 2014; Yoschenko et al., 2006). The Savannah River Site (SRS), which is located along the Savannah River in western

South Carolina, was constructed for the production and storage of various nuclear materials in the 1950s. During that time, release of radionuclides into the environment from stack emissions and cooling water discharge to surface waters have resulted in the incorporation of radionuclides in forest soils at SRS (Beals and Hayes, 1995a; Hejl et al., 2013; Paller et al., 2014; Xu et al., 2011). Despite the cessation of nuclear materials production and the continuation of site clean-up efforts, some concerns remain about the levels of radionuclides in the environment and their potential for transport, creating additional exposure pathways (Beals and Hayes, 1995b; Dong et al., 2012; Paller et al., 2014; Xu et al., 2011). Consequently, there is continuous radiological monitoring of the water, air, flora and fauna at SRS so as to

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ensure the safety of on-site workers and communities nearby SRS (Beals and Hayes, 1995a).

Prior to the cessation of production of most nuclear materials at SRS in 1991 (Commodore et al., 2012), stream systems at the site were contaminated by radionuclides through discharged cooling water from production facilities (Hiergesell and Phifer, 2014; Mackay et al., 2013). The radionuclides in the discharges, which was predominated by ^{137}Cs —a nuclear fission product, became bound in sediments and soil along wetland riparian forest downstream from reactor discharge points (Paller et al., 2008, 2014). Radioactive elements can be absorbed into the forest vegetation through root uptake, aerial deposition on foliage, and surface adhesion of re-suspended radionuclides on leaves and stems. For example, ^{137}Cs and ^{90}Sr , which are radionuclides with relatively long half-life (30.17 and 28.79 years, respectively), can be readily taken up by plants due to their high biological availability (Broadley and White, 2012; Kanter et al., 2010; Malek et al., 2002; Savinkov et al., 2007; Zhu and Smolders, 2000). Furthermore, the radionuclides can be cycled back to the soil from the vegetation through annual litter and periodic tree/branch fall creating contaminated wildland fire fuel. Thus, soil and vegetation contamination with radionuclides discharged from the facilities have a long-term radiological impact on the environment at SRS.

In this study, concentrations of radionuclides (i.e. fission products, and actinides and their decay products) were measured in surface soil and un-decayed vegetative matter (litter) samples collected along the stretches of streams that are part of the riparian ecosystem at SRS. The specific objectives of the study are to 1) investigate the distribution of anthropogenic and naturally occurring radionuclides between the riparian soil and litter of the forest along the selected streams in SRS, and discuss the factors that might influence this distribution; 2) investigate the difference in the activity concentrations of radionuclides in the soil/litter across the four streams, and determine the radionuclides that are the substantial contributors to radioactivity in SRS environment; and 3) determine the relationship between soil and litter radionuclide concentrations along the bank of the streams.

2. Materials and methods

2.1. Study site and sampling locations

SRS, which is located near Aiken, SC and administered by the United States Department of Energy, was established in the early 1950s for the production of nuclear materials. Prior to the cessation of the production of most nuclear materials in 1991, tritium and other radionuclides were released into the environment. Four of the five stream systems that drain the site received cooling water discharges from nuclear production related facilities. All the streams flow into the Savannah River which borders the site to the south and west. The four stream systems (Fourmile Branch, Lower Three Runs, Pen Branch, and Steel Creek) with historical inflow from nuclear facilities drain into the river through a swamp system (Fig. 1).

Surface soil and litter samples were collected from pairs of plots from 12 locations (48 plots in total) downstream from sources of radionuclides discharge/inflow into the streams (Fig. 2). The plots, starting from a location closest to the facilities, were systematically located along the stretch of each of the four streams. The plots were geospatially located in February 2013 using LiDAR derived topography map from 2009, digital ortho-photo data from 2010, road maps for access points, and gamma overflight (aerial radiological survey conducted by EG & G Energy Measurements Remote Sensing Laboratory, Bechtel, NV) data that was collected in 1998. The overflight signal primarily measures ^{137}Cs contamination levels in surface soil. The chosen locations were field checked from February 25th to March 11th, 2013 for forest type and accessibility, and were validated on the ground for gamma emission levels using an Electra Plus portable survey instrument (NE Technology Model Electra Plus with Alpha/Beta-Gamma

Detector). Some locations were moved if inconsistent gamma readings were measured or poor access existed. Each stream segment had sample locations on both sides due to access constraints, but both (paired) plots used for sampling were on the same side. The paired plots comprised of one plot in a contaminated area and the other in an uncontaminated area. Contaminated sample locations were selected in surface areas that held the highest gamma level readings, while corresponding non-contaminated background sample locations held levels below the minimum ambient instrument background. All gamma readings were less than Ambient Instrument Background (about 2500 disintegrations per minute (dpm)) for non-contaminated locations and above Ambient Instrument Background for contaminated locations (between ambient background and up to 8000 dpm).

2.2. Soil and litter sample collections

The sample collection started on March 11th, 2013 and was completed during the week of April 23rd, 2013. Each individual plot consisted of a 20×20 meter area in which five sub-plots were sampled for surface litter and soil. Gamma readings were used to confirm presence of contaminants at each sample point. A one square meter sample frame was used at each of the five sub-plots to obtain hardwood-pine leaf litter. No decayed litter was collected since the layer is normally very thin in riparian hardwood-pine forests and the decayed material does not contribute to the flaming front emissions. The flaming front is the zone of fire edge where the solid combustion is sustained and it is the major contributor to wildfire smoke emission. Litter samples were placed in plastic storage bags, sealed onsite, tagged and placed in a transportation container. A corresponding mineral soil sample below the decayed litter layer was collected from 0 to 15 centimeter (0–6 inch) depth with a spade shovel, composited into one plastic storage bag, sealed on-site, tagged and placed in an appropriate transportation container. Both soil and litter samples were weighed prior to drying upon sample-receipt and again after drying. All aliquots and percent moisture were tracked in Laboratory Information and Management System at the SRS Environmental and Bioassay Laboratory. Environmental Laboratory methods unique to SRS were instituted in analyzing the litter and soil samples. These methods have been published and recognized nationally (Maxwell et al., 2015a, 2010, 2015b).

2.3. Laboratory analysis

2.3.1. Pretreatment of litter samples

The detailed pretreatment of litter samples was well documented in a previous study (Viner et al., 2018). Briefly, litter samples were blended to create a homogeneous mixture after completely dried in the oven overnight. The sample mixed with standards in 250 mL zirconium crucibles were heated twice in muffle furnace to create white ash solids. After cooling, the sample was fused by adding sodium hydroxide. Water was then added to dissolve this fusion cake. The sample was transferred to a 225 mL centrifuge tubes following completely dissolved in water. After rinsing with 6M nitric acid, ferric nitrate (iron carrier) and lanthanum nitrate were added to the samples in the centrifuge tubes and the samples were diluted with DI water and cooled to room temperature. Ammonium phosphate, calcium nitrate, and titanium chloride were next added to the samples and mixed well. Hydrogen chloride was used to dissolve iron hydroxide precipitate that was formed following centrifuge and discarding supernatant. Hydrofluoric acid was added next, the samples were capped and mixed, then cooled briefly in ice bath for 15 min. Samples were centrifuged again and the supernatant was poured off. After a three-minute wait, sodium nitrite was added to oxidize Pu to Pu (IV). After concentrated nitric acid was added to this sample load solution, actinides were then separated using Eichrom (Lisle, IL) TEVA™, TRU™, and DGA™ resins into three fractions. Stacking of TEVA™ + TRU™ resin was then used to separate Th and U isotopes. Strontium resin was then used for the separation of $^{89/90}\text{Sr}$ from the

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