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Predicted cumulative dose to firefighters and the offsite public from natural and anthropogenic radionuclides in smoke from wildland fires at the Savannah River Site, South Carolina USA

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ABSTRACT

The contaminated ground surface at Savannah River Site (SRS) is a result of the decades of work that has been performed maintaining the country's nuclear stockpile and performing research and development on nuclear materials. The volatilization of radionuclides during wildfire results in airborne particles that are dispersed within the smoke plume and may result in doses to downwind firefighters and the public. To better understand the risk that these smoke plumes present, we have characterized four regions at SRS in terms of their fuel characteristics and radiological contamination on the ground. Combined with general meteorological conditions describing typical and extreme burn conditions, we have simulated potential fires in these regions and predicted the potential radiological dose that could be received by firefighting personnel and the public surrounding the SRS. In all cases, the predicted cumulative dose was a small percent of the US Department of Energy regulatory limit (0.25 mSv). These predictions were conservative and assumed that firefighters would be exposed for the duration of their shift and the public would be exposed for the entire day over the duration of the burn. Realistically, firefighters routinely rotate off the firefront during their shift and the public would likely remain indoors much of the day. However, we show that even under worst-case conditions the regulatory limits are not exceeded. We can infer that the risks associated with wildfires would not be expected to cause cumulative doses above the level of concern to either responding personnel or the offsite public.

1. Introduction

Wildfires can volatilize both natural and anthropogenic radionuclides. Man-made radionuclides in wildland fuels contaminated by nuclear releases are of great concern (Pazukhin et al., 2004; Hao et al., 2009). These radionuclides can be present in concentrated amounts due to aerial transport and deposition or as the result of releases to surface waters that are subsequently re-distributed in wetlands and forests downstream. The typical anthropogenic radionuclides of concern include radioisotopes of cesium (^{134,137}Cs), plutonium ($^{238/239}$Pu), uranium (^{234,235}U), and strontium ($^{89/90}$Sr). The most common anthropogenic contaminant found in the environment is usually ^{137}Cs (Paller et al., 2014). This is because ^{137}Cs has a relatively long physical half-life of 30.2 years, a high fission yield, and a high bioavailability-due to its physiological similarity to potassium. Certain natural radionuclides and their daughter products such as polonium (^{210}Po), radium (^{226,228}Ra), uranium (^{233,234,238}U), thorium (^{228,230,232}Th), lead (^{210,212}Pb), beryllium (^{7}Be), and potassium (^{4} K) are found in surface fuels throughout the world as a result of natural processes following cosmic ray interactions with particles in the atmosphere and decay of primordial geologic elements such as thorium and uranium (Sugihara et al., 1999; Persson and Holm, 2011; Hejl et al., 2013). Radionuclides can bio-accumulate in vegetation and then be re-cycled through litter fall to form components of the dead and live fire fuels. The natural radionuclides of greatest concern are those that are 1) common and long-lived, 2) easily volatilized, 3) have high energy particle emissions which can damage tissues, and 4) can accumulate in soft tissue (e.g. ^{210}Po) and bones (e.g. ^{226}Ra).

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In general, in most parts of the world, the individual natural radionuclides and man-made radionuclides from fall-out exist in the environment at concentrations too low to be a health risk relative to particulate matter (Paatero et al., 2009). However, during wildfires and prescribed fires, large quantities of radionuclides may be volatilized, or become attached to airborne particulates by the combustion process, and are detectable in smoke (Commodore et al., 2012; Volkerding, 2003). When radionuclides are considered individually, the concentration thresholds for a dose that exceeds the established worker limits are uncommon except for extreme values observed in wildland fuels contaminated in nuclear incidents (Viner et al., 2015). To date, no assessment has been made of cumulative dose to firefighters or to the public from natural radionuclides in conjunction with anthropogenic radionuclides from wildfires.

A method was recently developed to model emission, exposure, and dose to firefighters for any individual radionuclide during fires knowing only certain basic terms like fuel load and consumption, fire spread, the properties of the radionuclides, and their concentrations in the fuel (Viner et al., 2015). These components can then be coupled to a firefighter's physical location on the fire line, breathing rate, and shift length. The potential dose to a firefighter or a member of the public is a consequence of the cumulative dose from all radionuclides released during a fire, which can be calculated using this method. However, worldwide measurements contain very few observations of more than one radionuclide in wildland fuels either man-made or natural (Hejl et al., 2013; Viner et al., 2015). No published observations exist of a complement of man-made and dominant natural radionuclides in wildland fuels from which cumulative dose can be estimated. Our objective is to determine cumulative dose from both naturally occurring and man-made radionuclides and their relative contribution to the total dose to firefighters. From these results, we can 1) test the hypothesis that naturally occurring radionuclides are relatively small contributors to potential doses and 2) determine how fire dynamics may influence cumulative radiological concentrations in smoke and potential doses to firefighters and the public.

2. Methods

2.1. Forest and fire history

We estimate the potential doses to onsite firefighters and to the offsite public during wildfires for both anthropogenic and natural radionuclides present in forest surface fuels in areas contaminated from cooling water discharges at the Savannah River Site (SRS), South Carolina, USA. SRS is a large (800 km^2) U.S. Department of Energy nuclear facility located in the southeastern USA and it was built in the 1950's as part of the U.S. Cold War effort (Kilgo and Blake, 2005). During SRS's operational history, four major riparian or floodplain zones that flow through the Site were contaminated with various radionuclides as a result of discharges from nuclear processing facilities (Carlton, 1998). The forests that occupy the riparian areas of Fourmile Branch, Pen Branch, Steele Creek and Lower Three Runs were the focus of this study (Fig. 1).

The affected forest contains primarily loblolly pine (*Pinus taeda* L.), with some mixed hardwoods that include oaks (*Quercus* spp.), gums (*Nyssa* spp.), ash (*Fraxanus* sp.), and poplar (*Lirodendron* sp.). These riparian forests have been harvested sparingly since the Site was established in the early 1950's. However, from 1954 to 1988, portions of the flood plain forest vegetation were killed by hot water effluents from reactor facilities. These areas have either regenerated naturally over the last several decades or were planted (Barton et al., 2000). In these forests, the wildland fuels present to carry a fire are primarily litter, twigs, and branches. Live woody shrubs and grasses are present but are a small component of the available fuels to carry fires. No periodic prescribed burning has been conducted in these areas, except a single prescribed fire in 1994 in Pen Branch to clear selected areas prior to tree planting. The recent fire history of these areas is represented by occasional wildfires ignited by lightning. Fire control activities within these areas are limited because the soils are contaminated with radionuclides and disturbance is avoided to comply with the US Code of Federal Regulations (10 CFR 835). In the past, extensive wildfires have burned through the Savannah River swamp, adjacent flood plain and riparian forest during dry periods.

2.2. Radionuclide activity in fuels

The 1999 gamma spectrographic overflight (Aerial Radiological Survey by EG and G Energy Measurements, Bechtel Nevada, Remote Sensing Laboratory) for Fourmile Branch, Pen Branch, Steel Creek, and Lower Three Runs were overlaid with topographic information from a 2009 Light Detection and Ranging (LiDAR) overflight project and the most current aerial photos. The gamma overflight signal measures primarily surface ^{137}Cs contamination levels. We identified 48 paired sample points (contaminated, non-contaminated) in the study. Twelve systematically spaced sample locations along each of the four streams were located in February 2013 (Fig. 2). At each location, the contaminated sample point was confirmed with a surface radiological scanner using an Electra Plus portable survey instrument (NE Technology Model Electra Plus with Alpha-Beta-Gamma Detector), such that the observed gamma activity was the highest in the local vicinity. It was then marked and mapped using a global positioning system estimate. An adjacent non-contaminated paired sample point was similarly confirmed at the nearest point away from the stream such that the forest conditions, soils and topography were similar. It was then marked and mapped. For non-contaminated sampling locations, all field survey readings were less than ambient instrument background (about 2500 dpm). For the selected contaminated sampling locations, the field survey readings were between ambient background and up to 8000 dpm. We used sampling procedures similar to previous studies on SRS for wildland fuels to obtain a sufficient mass of surface fuel material of litter, small twigs, and branches (Maier et al., 2004). Beginning in March and ending in April 2013, four subsamples were obtained around each sample point from a 1 m^2 area plus an additional sample was taken at the central point. The material was placed in sample bags, labeled and sent to the SRS Environmental and Bioassay Laboratory for analysis. The composited vegetation fuel sample was dried in a 105 °C oven overnight until completely dry. The dried sample was then blended to create a homogeneous mixture. Each vegetation sample was analyzed for the reported radionuclides shown in Table 1 using the associated radioanalysis. An environmental laboratory method unique to SRS was instituted in analyzing the vegetation samples. This method has been published and recognized nationally (Maxwell et al., 2010).

For actinide analysis of vegetation fuel samples, an aliquot of 10–20 g of the dried and blended sample was measured into 250 mL zirconium crucibles. Standards were added to adjust for recovery efficiencies during processing. Crucibles were briefly dried on the hotplate and samples were then placed in a 200 °C muffle furnace and ramped to 600 °C for 2–4 h. Samples were removed from the furnace and allowed to cool. Concentrated nitric acid and hydrogen peroxide was added, 5 mLs each, and samples were carefully evaporated to dryness on a hotplate. Crucibles were placed back in the 600 °C furnace for 5–10 min or until the ash solids were white. Crucibles were then removed from the furnace and allowed to cool. Once cool, 15 g of sodium hydroxide was used for the fusion. Samples were covered and fused for 15 min. After fusion, the crucibles were removed from the furnace and allowed to cool. Water was then added to dissolve this fusion cake and the samples were transferred to 225 mL centrifuge tubes. All solids were dissolved by adding more water and heat to ensure complete dissolution. A final crucible rinse with 6 M nitric acid further removed any actinides from the crucible. 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.