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Mechanical environmental transport of actinides and ^{137}Cs from an arid radioactive waste disposal site

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ABSTRACT

Aeolian and pluvial processes represent important mechanisms for the movement of actinides and fission products at the Earth's surface. Soil samples taken in the early 1970's near a Department of Energy radioactive waste disposal site (the Subsurface Disposal Area, SDA, located in southeastern Idaho) provide a case study for studying the mechanisms and characteristics of environmental actinide and ^{137}Cs transport in an arid environment. Multi-component mixing models suggest actinide contamination within 2.5 km of the SDA can be described by mixing between 2 distinct SDA end members and regional nuclear weapons fallout. The absence of chemical fractionation between ^{241}Am and $^{239+240}$Pu with depth for samples beyond the northeastern corner and lack of ^{241}Am in-growth over time (due to ^{241}Pu decay) suggest mechanical transport and mixing of discrete contaminated particles under arid conditions. Occasional samples northeast of the SDA (the direction of the prevailing winds) contain anomalously high concentrations of Pu with ^{240}Pu/^{239}Pu isotopic ratios statistically identical to those in the northeastern corner. Taken together, these data suggest flooding resulted in mechanical transport of contaminated particles into the area between the SDA and a flood containment dike in the northeastern corner, following which subsequent contamination spreading in the northeastern direction resulted from wind transport of discrete particles.

1. Introduction

Particulate transport is an important mechanism for the movement of actinides and fission products at the Earth's surface. Decades of atmospheric weapons testing and nuclear accidents such as Chernobyl and Fukushima have resulted in significant injections of these isotopes in particulate form into the troposphere (Child and Hotchicis, 2013; Holgye and Maly, 2000; Holgye et al., 2004; Van Pelt and Ketterer, 2013; Petrovic et al., 2013). In addition, a very recent incident at the only operating US geologic repository for radioactive waste, the Waste Isolation Pilot Plant (WIPP), has resulted in the transfer of particulate actinides from the facility located in the subsurface to the surrounding above-ground environment (Wald, 2014; Tollefson, 2014). From the perspective of long-term environmental stewardship, these events highlight the importance of understanding the mechanisms and characteristics of contaminant resuspension and redistribution via convective and diffusive atmospheric processes.

The Subsurface Disposal Area (SDA) at Idaho National Laboratory (INL) represents a case study in particulate actinide environmental transport. From 1954 to 1970 the SDA operated as a shallow land disposal site for transuranic and fission product waste. During this period more than 10^{16} Bq of ^{137}Cs, ^{241}Am, and Pu (collectively) were buried in shallow, unlined excavations (Markham et al., 1978; Payne, 2006; Arrenholz and Knight, 1991; Lenhard et al., 2004; INEL, 1995). Wastes disposed of at the SDA were highly diverse and susceptible to contaminant release; disposal containers (containing various solid and liquid radioactive material forms) ranged from cardboard boxes, wooden boxes/crates, and steel/metal canisters, to loose waste dumped directly into excavations without any containment (Payne, 2006; Becker, 2002).

Two flooding events occurred at this site in the 1960's. In the early 1970's a large number of soil samples surrounding the SDA

Abbreviations: SDA, Subsurface Disposal Area; INL, Idaho National Laboratory; TIMS, thermal ionization mass spectrometry.

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were analyzed for the actinides by Markham et al.; their study confirmed that actinide contamination from the SDA to the surrounding environment had occurred (Markham et al., 1978). As the highest levels of contamination were observed in areas of lower topography (primarily within a drainage dike on the immediate northeastern corner, see Fig. 1), Markham et al. hypothesized that contamination transport in the near field of the SDA resulted from flood water transport, while the more widely disperse, lower concentrations of actinides in the northeast–southwest direction were transported by the wind. Although in the early 1970's low level actinide contamination had been observed up to 2.5 km to the northeast of the SDA, by 1995 low level Pu contamination from the SDA had been observed more than 48 km to the northeast (Beasley et al., 1998).

Radiometric and mass spectrometric analyses of Am, Pu, and ^{137}Cs together can reveal insights into the predominant transport mechanisms at this site. Under aqueous (freshwater) conditions at the INL site both Pu and ^{137}Cs are highly sorbed to clays and small soil particles (Groenewold et al., 2005; Mincher et al., 2003, 2004), whereas Am is anticipated to be slightly more soluble and thus partition to a greater extent between the solid and aqueous phases (Groenewold et al., 2005). Thus, under aqueous transport conditions (such as those present during the floods) Pu and ^{137}Cs transport would be expected to be governed primarily by pluvial transport and deposition of contaminated particles, whereas Am could be transported both as a particulate and as a soluble species (and thus potentially fractionate from Pu and ^{137}Cs). Conversely, under arid conditions Am, Pu, and ^{137}Cs are anticipated to be in particulate form and thus transport of all three would be expected to be similar (e.g. transport via Aeolian processes) (Hulse et al., 1999).

In Markham's original work, a lack of statistical difference in the ^{241}Am/$^{239+240}$Pu activity ratios with depth was observed for samples taken in the drainage dike and farther afield; although ^{241}Pu was not measured directly during this study, Markham et al. hypothesized that ^{241}Pu was preferentially associated with upper (0–4 cm) soil fractions and causing a localized in-growth of ^{241}Am. Importantly, this hypothesis is based on the assumption that Am is more prone to downward migration than Pu in the environment around the SDA, such as might occur under the aqueous conditions present during the flood (Groenewold et al., 2005). Interestingly, a study more than 40 years later by Payne et al. on soils from 2 locations in the drainage ditch showed significant ^{241}Am in-growth at only one of the locations (Payne, 2006; Payne et al., 2008), and that at both locations ^{241}Pu was distributed relatively equally between both 0–4 cm and 4–8 cm soil fractions (Payne, 2006). However, the study of Payne et al. analyzed soils from only the two locations with the highest contamination within the drainage ditch; to our knowledge no studies have focused on ^{241}Pu contamination from this event as a function of distance from the SDA. Analysis of the Pu/Am activity and isotope ratios in samples at distance could assist with identifying whether transport at a given location occurred predominantly under aqueous versus arid conditions.

Tracking particulate contamination from a given source relies on the ability to correlate the isotopic ratios of the element(s) of interest at a given location to those of the emitting source. Multi-component mixing models are one tool that is frequently utilized to perform these calculations, however such models require a priori knowledge of the isotopic composition of each end member. As more than 230 different processes have been identified as contributors to the waste within the SDA, the isotopic composition of any waste that may have been transported from the SDA is suspected to be highly diverse (Beasley et al., 1998).

One possible alternate technique to multi-component mixing models is the usage of ^{137}Cs as a surrogate for fallout Pu. As both Cs and Pu have similar environmental mobilities under both aqueous

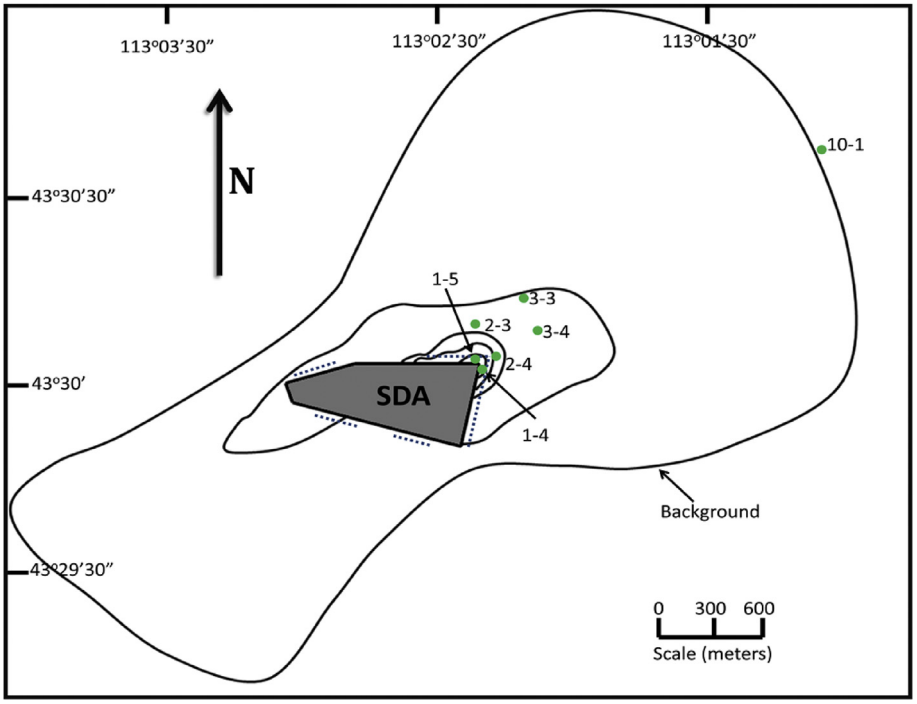

Fig. 1. Sampling locations surrounding the SDA. Note that 1-4 and 1-5 represent locations within a flood drainage dike; soil samples at these locations were disturbed during flooding and subsequent dike enlargement activities, while other sampling locations are reported to represent undisturbed locations (Markham et al., 1978). Solid black lines represent isopleth lines for contamination levels of 1300, 400, 25, and 5 nCi/m^2, decreasing with distance from the northeastern corner of the SDA (adapted from Markham et al., 1978). Dashed blue lines represent the location of the drainage ditch built after the 1962 flood. Green dots represent soil sampling locations. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)