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Plutonium in wildlife and soils at the Maralinga legacy site: persistence over decadal time scales

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ABSTRACT

The mobility of plutonium (Pu) in soils, and its uptake into a range of wildlife, were examined using recent and ~25 year old data from the Taranaki area of the former Maralinga weapons test site, Australia. Since its initial deposition in the early 1960s, the dispersed Pu has been incorporated into the soil profile and food chain through natural processes, allowing for the study of Pu sequestration and dynamics in relatively undisturbed semi-arid conditions.

The data indicate downward mobility of Pu in soil at rates of ~0.2–0.3 cm per year for the most mobile fraction. As a result, while all of the Pu was initially deposited on the ground surface, approximately 93% and 62% remained in the top 0–2 cm depth after 25- and 50-years respectively. No large-scale lateral spreading of the Taranaki plume was observed. Pu activity concentrations in 0–1 cm soils with biotic crusts were not elevated when compared with nearby bare soils, although a small number of individual data suggest retention of Pu-containing particles may be occurring in some biotic crusts.

Soil-to-animal transfer, as measured by concentration ratios ($CR_{wo\text{-}soil}$), was 4.1E−04 (Geometric Mean (GM)) in mammals, which aligns well with those from similar species and conditions (such as the Nevada Test Site, US), but are lower than the GM of the international mammal data reported in the Wildlife Transfer Database (WTD). These lower values are likely due to the presence of a low-soluble, particulate form of the Pu in Maralinga soils. Arthropod concentration ratios (3.1E−03 GM), were similar to those from Rocky Flats, US, while values for reptiles (2.0E−02 GM) were higher than the WTD GM value which was dominated by data from Chernobyl. Comparison of uptake data spanning approximately 30 years indicates no decrease over time for mammals, and a potential increase for reptiles. The results confirm the persistence of bioavailable Pu after more than 50 years since deposition, and also the presence of larger-sized particles which currently affect $CR_{wo\text{-}soil}$ calculations, and which may serve as an ongoing source of bioavailable Pu as they are subjected to weathering into the future.

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1. Introduction

While many past studies have focused on pathways of human exposure (e.g., ANSTO, 1990), recent emphasis on the need to assess ionising radiation exposure of wildlife has rekindled research interest in the occurrence, mobilisation, and uptake of radionuclides (Howard et al., 2013). Uptake of radionuclides by wildlife is typically quantified as a concentration ratio ($CR_{wo\text{-}media}$) between the organism whole-body activity concentration and the activity concentration in an environmental medium (Beresford, 2010; Beresford et al., 2008; Howard et al., 2013). For terrestrial systems:

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$$CR_{wo-soil} = \frac{\text{activity concentration in organism whole} - \text{body (fresh mass)}}{\text{activity concentration in soil (dry mass)}}$$

The $CR_{wo\text{-}media}$ parameter aggregates complex biotic and abiotic factors affecting uptake, including how the organism becomes exposed to the contaminant, which is typically distributed unevenly in soils and is subject to change over time. For example, Pu deposited on soil typically exhibits a decreasing activity concentration with depth. This can lead to greater exposure to organisms at the soil surface compared with organisms feeding or burrowing below ground. However, Pu can migrate downward into soils (Kersting, 2013; Sakaguchi et al., 2004; Tims et al., 2013) and thus change exposure and uptake over time for organisms accessing particular soil strata. Similarly, an inhomogeneous spatial distribution of a radionuclide in specific types of environments (e.g. biotic soil crusts) may affect wildlife differently according to how they use and interact with these environments. For these reasons, a thorough investigation of soil-to-wildlife transfer requires better understanding of how contaminants are distributed in soil, and how such distributions change over time.

Plutonium (Pu) isotopes can be relatively important contributors to radiological dose to wildlife when internalised within the body (Vives i Batlle et al., 2011). At certain weapons testing sites, such as Area 13 (Nevada Test Site, US) and Taranaki (Maralinga, Australia), the Pu contamination was dispersed and deposited onto the nearby land surface during explosive sub-fission or partial-fission tests. Although remedial actions have been conducted (e.g., MARTAC, 2003), residual Pu remains in areas of these sites at activity concentrations that are elevated above background levels. Given the elapsed time since deposition (circa 1950s and 60s), there now exists an opportunity to evaluate Pu sequestration, mobility, and biotic uptake following its incorporation into ecosystem soils and biotic media; and to compare data over decadal time scales.

Given the low solubility of most Pu forms, its mobility in the environment is typically associated with erosion, transport and transfer of Pu-containing compounds, colloids and larger particles to which Pu has adhered (Kersting, 2013). Transport rates may be inhibited by environmental features such as biotic crusts on desert soils which resist erosion, although data are generally lacking (ANSTO, 1998; Gilbert et al., 1988).

Over long time frames (>100 s of years), the mobilisation rates of particle-adsorbed radionuclides are not expected to be constant but rather subject to infrequent episodes of extreme wind, flooding or landscape fire events that can dominate the eventual transport rates (Breshears et al., 2003). Additional changes over time to the bioavailable fraction can result from processes such as weathering of particles (Salbu, 2001) and increased solubilisation/mobilisation due to biotransformation of Pu forms (Francis, 2001). However, long-term mobility studies of actinides are generally lacking in the scientific literature, largely due to the difficulty of taking measurements over the time scales required. Such studies are now becoming feasible at selected sites such as Taranaki, Maralinga, which have data available from the testing period (1950s–60s), earlier studies in the 1980s, and now more recent study at 50-years post-deposition.

A key means of monitoring changes in Pu distributions in the environment over time is the use of isotopic ratios, which can be used to identify a specific contamination source and aid in monitoring the spatial distribution of the contamination from that source. Although some Pu isotopic ratios of the Maralinga test site are available, (ARL, 1988; Child and Hotchkis, 2013), data have not been published on the specific Pu isotope ratios of most of the seven sites used for nuclear detonations at Maralinga. Previous measurements were conducted using gamma spectrometry where the presence of ^{152}Eu interfered with the sensitivity of the ^{240}Pu measurement. Determining ratios for the previously unmeasured sites as well as confirming the ratios given by gamma spectrometry analysis will allow for better future assessment of changes in Pu occurrence and distribution.

The objectives of this study were: to better quantify Pu distributions in soils, including soils with biotic crusts; to determine the persistence and mobility of Pu over 25-, and 50-year time scales; and to increase understanding of Pu uptake in a range of mammal, reptile and arthropod wildlife including comparison with $CR_{wo\text{-}soil}$ values from similar sites and the current international database (Wildlife Transfer Database (WTD), www.wildlifetransferdatabase.org) (Copplestone, in press).

2. Materials and methods

2.1. Site description and Pu contamination history

The Maralinga test site is located on the southern edge of the Great Victoria Desert, South Australia (Fig. 1). Semi-arid conditions exist with sparse and erratic rainfall (annual mean of 224 mm). The ground surface is relatively flat and given the highly transmissive (sandy) soils, there is minimal evidence of reworking/transport of soils by surface water. Monthly mean temperatures range from

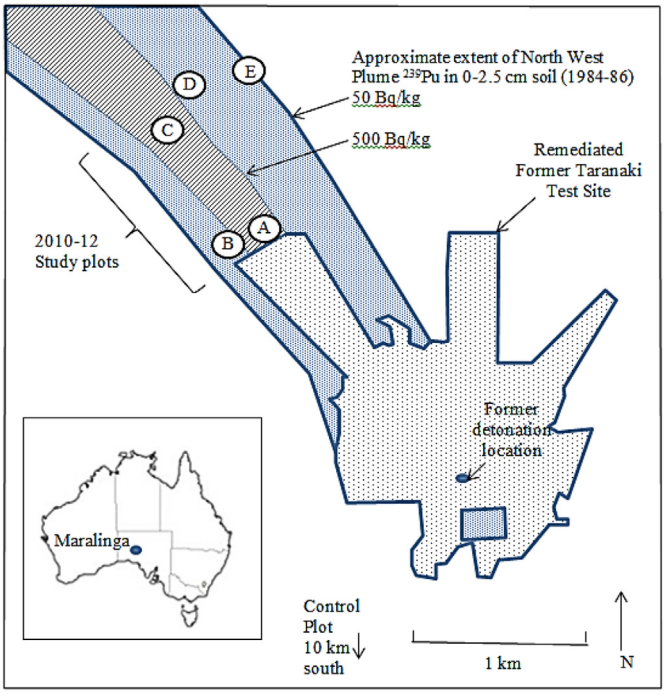

Fig. 1. Study plots and NW Plume relative to the remediated former Taranaki Test Site, Maralinga, South Australia.

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