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Measurement of fallout radionuclides, ^{239,240}Pu and ^{137}Cs, in soil and creek sediment: Sydney Basin, Australia

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ABSTRACT

Soil and sediment samples from the Sydney basin were measured to ascertain fallout radionuclide activity concentrations and atom ratios. Caesium-137 (^{137}Cs) was measured using gamma spectroscopy, and plutonium isotopes (^{239}Pu and ^{240}Pu) were quantified using accelerator mass spectrometry (AMS).

Fallout radionuclide activity concentrations were variable ranging from 0.6 to 26.1 Bq/kg for ^{137}Cs and 0.02–0.52 Bq/kg for $^{239+240}$Pu. Radionuclides in creek sediment samples were an order of magnitude lower than in soils. ^{137}Cs and $^{239+240}$Pu activity concentration in soils were well correlated ($r^2 = 0.80$) although some deviation was observed in samples collected at higher elevations. Soil ratios of ^{137}Cs/$^{239+240}$Pu (decay corrected to 1/1/2014) ranged from 11.5 to 52.1 (average = 37.0 ± 12.4) and showed more variability than previous studies.

^{240}Pu/^{239}Pu atom ratios ranged from 0.117 to 0.165 with an average of 0.146 (±0.013) and an error weighted mean of 0.138 (±0.001). These ratios are lower than a previously reported ratio for Sydney, and lower than the global average. However, these ratios are similar to those reported for other sites within Australia that are located away from former weapons testing sites and indicate that atom ratio measurements from other parts of the world are unlikely to be applicable to the Australian context.

1. Introduction

Atmospheric testing of atomic weapons in the 1950s and 1960s dispersed anthropogenic radionuclides across the globe (UNSCEAR, 2000). With a combined explosive yield of 440 Mt, this testing resulted in several tonnes of plutonium being deposited as fallout as well as numerous other radionuclides, including the fission product ^{137}Cs.

Nuclear tests were undertaken in the South Pacific region, including 12 in Australia. The testing in Australia, undertaken by the British Government, commenced in 1952 at the Montebello Islands off the coast of Western Australia with operation "Hurricane" (Symonds, 1985). In 1953, "Totem 1" and "Totem 2" were carried out at Emu Field in South Australia (Symonds, 1985). The "Mosaic" tests, comprising two detonations (G1 and G2) with the second having a ~100 kt yield (Child and Hotchkis, 2013), were carried out at the Montebello Islands in May and June 1956 (Butement et al., 1957; Symonds, 1985). Further testing was then carried out at Maralinga in South Australia where the "Buffalo" series, comprising four weapons, were detonated during September and October 1956 (Butement et al., 1958; Symonds, 1985) and the "Antler" series, comprising three tests, carried out during September and October 1957 (Dwyer et al., 1959; Symonds, 1985). The combined yield of the 12 tests carried out in Australia was approximately 220 kt with the largest single yield from the Mosaic-G2 test (Tims et al., 2013).

Additionally, a number of other tests termed the "Minor Trials" and "Assessment Trials" took place at Emu Field in 1953 and at Maralinga between 1955 and 1963 to improve explosive yield and assess the performance of weapon components, including under accident scenarios (Symonds, 1985) which involved burning and conventional explosive dispersal of plutonium, uranium and other radionuclides (Long et al., 2004). The minor and assessment trials at Emu Field and Maralinga were code-named "Kittens", "Tims", "Rats" and "Vixen" and were eventually given the title "Maralinga Experimental Programme" (Symonds, 1985).

'Long range' or 'delayed' fallout is comprised of fine radioactive

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aerosol particles from the fission reaction and those formed by the vaporisation of un-fissioned radioactive material, bomb casing and ancillary equipment (Butement et al., 1957). 'Close in' or 'short range' fallout is produced when updraughts carry a wide range of particles that have become radioactive by close contact with fission products (Butement et al., 1957). The relatively heavy particles settle out more quickly and are deposited closest to the test site, while the smaller particles generally travel further and may contribute to fallout at regional or hemispheric scales. Some aerosols, propelled into the atmosphere, may drift around the earth several times before settling out (Butement et al., 1957).

During the 1950s and 1960s, beta radioactivity measurements were made at stations located across Australia using gummed film exposed for 24 h periods (Dwyer et al., 1957). Beta radioactivity was detected at locations on the east coast of Australia, including Sydney (a major population centre), following the Grapple trials at Christmas Island (Dwyer et al., 1957), the Buffalo (Butement et al., 1958) and Antler (Dwyer et al., 1959) trials in Australia as well as the later testing carried out by France in Polynesia (Gibbs et al., 1967, 1968, 1969). Some radioactivity detected was attributed to global fallout (Alsop et al., 1963; Blake et al., 1960, 1962).

During the 1970s, $^{240}Pu/^{239}Pu$ atom ratios were measured in samples from Australia as part of a global study, which found that the global mean $^{240}Pu/^{239}Pu$ atom ratio was 0.176 (±0.014) (Krey et al., 1976). However, there was latitudinal variation in this atom ratio, and a later study by Kelley et al. (1999) proposed a higher mean ratio of 0.185 (±0.047) in the mid-latitudes of the Southern Hemisphere (30–53° S). Despite this, two sites on the east coast of Australia have shown slightly lower $^{240}Pu/^{239}Pu$ ratios of 0.1768 (±0.0027) (Brisbane, 27° S) and 0.1716 (±0.0014) (Melbourne 37° S) (Kelley et al., 1999). The same sites, measured earlier by Krey et al. (1976) had $^{240}Pu/^{239}Pu$ ratios of 0.1683 (±0.002) (Brisbane) and 0.1720 (±0.005) (Melbourne) that were also below the southern (and global) mean. The Krey et al. (1976) study also measured soil from other sites in Australia, including Sydney, where the $^{240}Pu/^{239}Pu$ atom ratio was found to be 0.1707 (±0.001). These measurements, albeit from a small sample set, suggested that the atom ratio on the east coast of Australia may be lower than the global average.

Recent studies across continental Australia (Child and Hotchkis, 2013; Everett et al., 2008; Tims et al., 2013) confirmed that $^{240}Pu/^{239}Pu$ isotopic ratios in Australia are generally lower than the global average, and in some areas of the country are significantly lower than the values originally determined for the three east coast Australian cities (Brisbane, Melbourne and Sydney). Soils in close proximity to former weapons test sites have extremely low $^{240}Pu/^{239}Pu$ isotopic ratio values (0.01–0.025 Emu Field, South Australia (Child and Hotchkis, 2013); 0.05–0.07 Taranaki, Maralinga, South Australia (Johansen et al., 2015); and 0.046–0.053 Monte Bello Islands (Child and Hotchkis, 2013)) which Child and Hotchkis (2013) suggest is indicative of low yield devices and possibly short irradiation times during weapon material production. With increasing distance from the test sites, these ratios approach typical global fallout values due to a decreasing contribution of low atom ratio material from the test sites. However, in most cases atom ratios away from test sites are lower than global fallout, indicating there may be a broader regional influence on $^{240}Pu/^{239}Pu$ atom ratios from testing conducted in Australia.

$^{239,240}Pu$ and/or ^{137}Cs have been used in recent (e.g. past 60 years) erosional studies (Everett et al., 2008; Hoo et al., 2011; Simms et al., 2008; Smith et al., 2012) and as a chronometer (Appleby, 2001; Crusius and Anderson, 1995; Hancock et al., 2011). ^{137}Cs is used as a tracer due to its low mobility, which is largely a consequence of its tendency to be adsorbed by small soil particles through the cation exchange sites on the clay mineral surfaces (Everett et al., 2008). Pu has a more complicated chemistry than Cs, experiencing multiple oxidation states and variable sorption as influenced by the presence of fulvic and humic acids (Fujikawa et al., 1999). Plutonium has also been found to be more particle reactive than Cs, binding to hydrous oxide soil coatings (Alberts and Muller, 1979).

Historically, ^{137}Cs (t ½ = 30.08 years (Browne and Tuli, 2007)) was favoured in these studies as it is easily detected by the non-destructive gamma-ray spectroscopy technique. However nearly two half-lives have elapsed since the peak release of ^{137}Cs in 1963 and activity concentrations are becoming more difficult to quantify. Plutonium isotopes have much greater half-lives (^{239}Pu t½ = 24110 years (Browne and Tuli, 2014) and ^{240}Pu t½ = 6561 years (Browne and Tuli, 2006)) and are becoming commonly used for erosional and sedimentary studies. A better understanding of the regional variability of Pu activity concentrations could benefit soil tracer studies while a better understanding of the atom ratio variability may assist with chronometric applications and fingerprinting Pu sources.

Sensitive techniques such as accelerator mass spectrometry (AMS) are able to quantify these isotopes in small (<2 g) samples down to sub-femtogram concentrations, although the technique is destructive and requires labour-intensive chemical separation and purification prior to measurement. Sensitive mass spectrometric measurements of ^{239}Pu and ^{240}Pu enable the $^{240}Pu/^{239}Pu$ atom ratio to be determined. Alpha spectrometry, although less sensitive than some mass spectrometry techniques, can also be used to measure these isotopes of plutonium, however the alpha emission energies of ^{239}Pu and ^{240}Pu overlap and a combined $^{239+240}Pu$ activity concentration is determined. This technique also requires multiple chemical separation and purification steps and is similarly destructive.

The Sydney region is Australia's largest population centre and is the location of Australia's only nuclear research reactor, at the Lucas Heights Science and Technology Centre (LHSTC). Near the LHSTC is an associated legacy low level waste site, the Little Forest Legacy Site (LFLS), formerly known as the Little Forest Burial Ground (LFBG), where small amounts of plutonium and other radionuclides were disposed of in trenches in the 1960s (Payne, 2012; Payne et al., 2013). Environmental monitoring of the LHSTC and LFLS has been undertaken since 1959 (Giles and Stockdale, 1966) and 1966 (Cook et al., 1969) respectively. During the years that fallout from weapons testing was being measured across Australia, radioactivity attributable to aerial dispersion from the LHSTC was not detected (Cook and Dudaitis, 1970a,b; Cook et al., 1969).

High volume air filters have been collected at LFLS since 1984 for the measurement of $^{239+240}Pu$ in airborne dust (Giles and Dudaitis, 1986). The values have been below minimum detectable activity (MDA), measured by alpha spectrometry, except for a possible trace reported in 1985 (Giles et al., 1988). Although not associated with atmospheric releases, $^{239+240}Pu$ has been detected in the soils at LFLS associated with soil–water transport (Payne et al., 2013).

The primary aim of the present study is to quantify the $^{239,240}Pu$ and ^{137}Cs activities as well as $^{240}Pu/^{239}Pu$ atom ratios in soil and creek sediment in the Sydney Basin, in order to establish general fallout values for this region. These data will supplement the growing number of $^{240}Pu/^{239}Pu$ atom ratios that have been collected for continental Australia. They will also provide baseline values for the Sydney region which may enable radionuclide detections in soils at the LFLS to be distinguished from local background. In addition, as experienced by countries around the world after the Chernobyl and Fukushima events, baseline data is important for quantifying the impact from any accidental releases from operational nuclear facilities, including those in neighbouring countries.

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