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Baseline studies of plutonium and caesium deposition in the Sydney Region

Brodie S. Smith

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Baseline studies of plutonium and caesium deposition in the Sydney Region

Abstract

This study presents an examination of fallout derived radioactivity in soil and alluvial sediment samples from the Sydney Basin. This work was undertaken to add to the growing dataset of published values on radioactivity from sites across Australia and to provide baseline data for the Sydney region. Such data provides further information on fallout patterns in Australia, and enables baseline values of radionuclides such as $^{240+239}\text{Pu}$ and ^{137}Cs to be established for the Sydney Basin in which Australia's major nuclear facility, with an associated legacy low-level waste site is located.

Samples were collected in an east-west transect across the Sydney Basin and Blue Mountains, resulting in the collection of 10 soil samples and 4 alluvial sediment samples. These were analysed by gamma spectroscopy for caesium-137 and with Accelerator Mass Spectroscopy for plutonium concentrations and $^{240/239}\text{Pu}$ isotopic ratios.

Plutonium-240+239 concentrations are found to be 0.24 Bq/kg (standard deviation 0.2, range 0.04-0.52 Bq/kg) for the <63 micron fraction. The caesium-137 activities averaged to 11.7 Bq/kg for soils and 2.1 Bq/kg for sediments.

Results from Plutonium-240/239 isotope studies agree with previous data indicating that continental Australia has a $^{240/239}\text{Pu}$ ratio that is lower than the global average. The average ratio of 0.15 (ranging from 0.11-0.16) for the Sydney Basin is found to be significantly lower than the accepted global mean of 0.176, indicating regional influences on Australian fallout. Similarly to previous studies, the Pu was concentrated preferentially in the finer soil fractions. Levels of caesium were approximately proportional to the plutonium, which suggests that Pu will be a suitable long-term substitute for caesium as an environmental tracer in Australia.

Radionuclide data from various soil and sediment samples collected at the legacy low-level waste site known as Little Forest Legacy Site (LFLS) indicates actinides such as isotopes of Pu are reaching the soil surface as a result of overflowing trenches during rainfall periods. This baseline data will allow for the disentanglement between the contributions of the legacy waste site and global fallout.

In order to explain the variations in caesium-137 and plutonium 240+239 between sites environmental factors were also measured. These include pH, rainfall, elevation, organic content as well as soil moisture. There was no correlation found between the measured environmental factors and the fallout activities.

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Brodie S Smith

A research report submitted in partial fulfilment of the
requirements for the award of the degree of
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The information in this thesis is entirely the result of investigations conducted by the author, unless otherwise acknowledged, and has not been submitted in part, or otherwise, for any other degree or qualification.

- Brodie Sabine Smith

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Abstract

This study presents an examination of fallout derived radioactivity in soil and alluvial sediment samples from the Sydney Basin. This work was undertaken to add to the growing dataset of published values on radioactivity from sites across Australia and to provide baseline data for the Sydney region. Such data provides further information on fallout patterns in Australia, and enables baseline values of radionuclides such as $^{240+239}\text{Pu}$ and ^{137}Cs to be established for the Sydney Basin in which Australia's major nuclear facility, with an associated legacy low-level waste site is located.

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Glossary of abbreviations

ADH – Australian Height Datum

AMS- Accelerator Mass Spectrometry

ANSTO- Australian Nuclear Science and Technology Organisation

ANTARES – Australian National Tandem Research Accelerator

BOM- Bureau of Meteorology

Cs- Caesium

GIS- Geographic Information Systems

HIFAR – High Flux Australian Reactor

ISO – International Organization for Standardization

LFBG- Little Forest Background Site

LFLS- Little Forest Legacy Site

LHSTC- Lucas Heights Science and Technology Centre

LOI- Loss on Ignition: test for organic content

NIST – National Institute of Standards and Technology

OPAL – Open Pool Australian Lightwater

PEG- polyethyleneglycol

Pu- Plutonium

TNT- trinitrotoluene

Introduction and study aims

Following atmospheric testing of atomic weapons in the 1950's and 1960's anthropogenic radionuclides were dispersed across the globe establishing a base level of contamination, to which any additional radioactive material will be added. With a combined explosive yield of 400Mt (UNSCEAR, 2000) atmospheric weapons testing resulted in several tons of plutonium being spread over the earth's surface. This was accompanied by numerous other fallout radionuclides, including the fission product ^{137}Cs (an isotope of caesium). The primary aim of this study is to quantify the plutonium and caesium deposited in the Sydney region as a result of nuclear weapons testing in order to establish a baseline to which other future radioactive contamination can be compared. The levels of fallout radionuclides are of interest for various reasons, including the potential long-term health impacts, studies of atmospheric mixing and circulation following the explosions, and in erosion studies. It is hoped that this study will increase the understanding of the spread of radionuclides resulting from the weapons testing allowing us to have a better understanding of patterns in global fallout.

Atmospheric Atomic testing was conducted in Australia by the United Kingdom beginning in 1952 at the Montebello Islands off Western Australia (named the Hurricane and Mosaic trials). The trials continued until 1958 with test sites located at Christmas Island (Grapple trials), Maralinga S.A (Buffalo and Antler trials) and at Emu Fields S.A (Totem tests). These test detonations were coordinated to coincide with atmospheric conditions which would limit the fallout that would be received by Australia. Conditions were chosen as to direct the fallout away from major population centres or in the case of the Montebello Islands out into the Indian Ocean. Despite these efforts fallout was recorded in Sydney following the Mosaic, Buffalo and Grapple trials (Butement et al., 1958, Butement et al., 1957, Dwyer et al., 1957, Keam et al., 1958). This was much lower than other areas of Australia, but is significant enough to allow for the assumption that fallout did reach the Sydney region, and that using modern highly sensitive equipment we will be able to measure it.

The Sydney region is home to Australia's only nuclear reactor, the OPAL reactor which is used for the production of radiopharmaceuticals and research. The region is also home to a legacy waste site. Both the presence of the reactor and the waste site make baseline studies of radionuclides important for the Sydney region as these pose possible sources of contamination.

The second aim of this study is to use the baseline data to assess the impact of the plutonium which was disposed in a legacy waste site associated with the former Atomic Energy Commission. Between 1960 and 1968 the Atomic Energy Commission (predecessor to ANSTO) disposed of its radioactive waste in a series of trenches across the road from its site at Lucas Heights, NSW. This site is now known as Little Forest Legacy Site (LFLS). The waste included several grams of plutonium along with other radionuclides. As a result of the poor hydro conductivity of the underlying geology of the trenches, it has been shown that in periods of high rainfall the trenches will fill with water, and radionuclides will overflow (Payne et al., 2013). This is a process that has been dubbed 'bath tubbing' or the 'bathtub effect'. (Payne et al., 2013). To investigate this bath tubbing surface soil samples have been

collected from the site. Samples close to the trenches have high levels of plutonium although Pu activity is reduced with distance from the trenches (Payne et al., 2013).

The establishment of a baseline for plutonium concentrations will enable the determinations of the extent of the contamination caused by the bath tubbing as well as distinguishing between trench plutonium and that of global fallout through the use of the $^{240/239}$ Pu ratio.

The third aim of this study is to add to the growing number of $^{240/239}$ Pu isotope ratios that have been collected for continental Australia. These values are indicative of the source of the plutonium. A global mean of ~0.18 has been accepted, though individual sites can differ significantly from this. Sites with low 240/239 values are usually the result of close in fallout from atomic testing. This consists of the larger particles that do not travel any great distance from the source, while higher results above the mean can be seen nuclear reactor sites such as Chernobyl (Child and Hotchkis, 2013). The determination of the $^{240/239}$ Pu isotope ratio will greatly increase our understanding of the atomic testing conducted here as well as having implications for erosion and sedimentation studies and geochronology.

Currently there are very few studies measuring the $^{240/239}$ Pu ratio as it requires highly sensitive equipment for detection. In the past Kelly (1999) and Krey (1976) have measured the plutonium- 240/239 ratio for Australia and have found it to be below Krey's accepted global mean of 0.176. More recently scientists from ANSTO and the Australian National University have used Accelerator Mass Spectrometry (AMS) in order to measure the ratios of sites in Australia (Child and Hotchkis, 2013, Everett et al., 2008, Tims et al., 2013). Tims (2013) measured $^{240/239}$ Pu isotopic ratios for multiple sites across continental Australia, concluding that the ratio is significantly below the global mean. While covering the majority of Australia this study only had one site from NSW, and it was located within the Snowy Mountains. As Sydney was missing from this continent wide study, it was decided that it was important to quantify the fallout in the region via the 240/239 ratio.

In order to achieve these aims, soil and alluvial sediment samples have been collected from sites in the Sydney Basin and Blue Mountains that are considered to be undisturbed since the atomic era of the 1950's and 1960's. These samples were then analysed using gamma spectrometry for caesium-137, Accelerator Mass Spectrometry for plutonium 240/239 ratios and plutonium 239+240 activity, and Alpha Spectrometry for plutonium 239+240 activities. In order to determine the causes of any variations within the activities measured a number of environmental factors were also measured including pH, organic content, elevation, rainfall, and soil moisture.

The study scope was limited to examining plutonium-240+239 and caesium-137 despite many other radionuclides being present from both the atomic testing and the Little Forest Legacy site. In addition, in order to provide a representative survey of radioactivity in the Sydney Basin within the limits of this nine month project, samples were collected in an east-west transect across the Sydney Basin.

It is expected that the results of this study will have a wide range of applications from public health monitoring to geomorphic research into erosion and deposition.

Chapter 1- Literature Review

1.1 Atomic testing in Australia

In order to assess the fallout received by the Sydney Basin it is necessary to investigate the sources of radionuclides in the Australian environment. Nuclear explosions can be described in terms of yield, with low yield weapons being the 20 kiloton weapons with an equivalent explosive power as 20 000 tons of TNT(trinitrotoluene) and high yield weapons having the explosive power of 20 megatons of TNT(Green 1962).

Kiloton devices are based on the explosive fusion of either uranium-235 or plutonium-239, the fission of which results in the formation of many isotopes. The most commonly studied of these are strontium-90, caesium-137, iodine-131, ruthenium-103, zirconium-95, carbon-14, barium-140 and cerium-144 (Green 1962).

There are more than one thousand different radionuclides released as the result of fission and neutron activation that occurs during the detonation of an atomic weapon, though many of these decay into their progeny's within minutes of the detonation (Kraus and Foster, 2014).

During the period of 1952-1958 the United Kingdom conducted several atomic tests both on the Australian mainland and on the Montebello Islands off the north-west coast of Western Australia. When these atomic weapons were detonated a large cloud of radioactive material was released. The fine components of this can travel in the atmosphere forming the long range or delayed fallout. This cloud may travel several times around the world, even following a narrow band of latitude, slowly losing radioactive material to the lower levels of the atmosphere(Keam et al., 1958). Here rain may quickly take the fallout to the ground. The location of deposition will depend on multiple climatic factors including wind direction and rainfall (Keam et al., 1958).

During all tests the close in fallout of the larger particles fell entirely within the exclusion zone (Butement et al., 1957). The combined yield of the testing was relatively small at ~220kT with the Maralinga tests contributing the most(~100kT from Mosaic G2) (Child and Hotchkis, 2013). Other experiments were also undertaken at Maralinga between 1959 and 1963, that involved the burning and dispersal of radionuclides (Tims et al., 2013). Table 1.1 contains information on atomic weapons testing conducted in Australia during the 1950's and 1960's.

Prior to the testing, the Australian Weapon Test Safety Committee set up a network of stations across the continent to measure fallout radioactivity. Testing at this time was undertaken using a piece of gummed paper (12 inches by 6 inches) which was left exposed for 24 hours before being sent via air mail to the X-ray and Radium Laboratory in Melbourne(Dwyer et al., 1957). These results of these readings can be seen in Table 1.1. As one would expect the ability to measure the radiation was limited in the 1950's with only total radioactivity available, not specific radionuclides. The units are presented as they were in the papers of the time. Only after the Mosaic G1 test was any radiation detected in the Sydney region and this was from an air filter sample(Butement et al., 1957). All the tests

presented are atmospheric (rather than underground) and will have contributed to the global fallout.

Table 1.1 Atomic testing undertaken by the United Kingdom in Australia during the 1950's and 60's (Geoscience, 2008, Butement et al., 1958, Butement et al., 1957, Dwyer et al., 1957, Keam et al., 1958).

| Date | Site | Name | Fallout to reach Sydney |
|-------------------|--------------------|-----------|---|
| 03/10/1952 | Monte Bello Island | Hurricane | |
| 14/10/1953 | Emu Fields | Totem 1 | |
| 26/10/1953 | Emu Fields | Totem 2 | |
| 16/05/1956 | Monte Bello Island | Mosaic G1 | Below detection limits using the gummed paper method. 0.02nCi/m ² using air filters |
| 19/06/1956 | Monte Bello Island | Mosaic G2 | Below detection limits |
| 27/09/1956 | Maralinga | Buffalo | Below detection limits |
| 04/10/1956 | Maralinga | Buffalo | 0.01 and 0.03 μ Ci/m ² |
| 11/10/1956 | Maralinga | Buffalo | 0.04 and 0.01 μ Ci/m ² |
| 22/10/1956 | Maralinga | Buffalo | |
| 15/05/1957 | Christmas Island | Grapple | 32mCi/km ² – using gummed paper method. |
| 31/05/1957 | Christmas Island | Grapple | |
| 19/06/1957 | Christmas Island | Grapple | |
| 14/09/1957 | Maralinga | Antler | |
| 25/09/1957 | Maralinga | Antler | |
| 09/10/1957 | Maralinga | Antler | |
| 08/11/1957 | Christmas Island | Grapple | |
| 28/04/1958 | Christmas Island | Grapple | |
| 22/08/1958 | Christmas Island | Grapple | |
| 02/09/1958 | Christmas Island | Grapple | |
| 11/09/1958 | Christmas Island | Grapple | |
| 23/09/1958 | Christmas Island | Grapple | |

Note 1 Ci = 3.7×10^{10} decays per second= approximately 37000000000 Bq

During the atomic testing of 1957, the stations around the country were measuring the total detectable activity using the gummed paper method. Following the Christmas Island trials a reading of 32 mCi/km² was measured for Sydney using the gummed paper. This reading was believed to originate from the Grapple 1 testing (Dwyer et al., 1957). These methods were once again used for the testing in 1958. For the Sydney region, a sample taken on the 7th June 1957 recorded a level an activity of 19 mCi/km² (Kear et al., 1958).

The measurements for Sydney were of a known age coming from the Buffalo trials. These readings were the largest detected for this set of trials (Kear et al., 1958). These historic studies indicate that fallout from the testing was reaching the Sydney region.

The tests were undertaken at points in time when the metrological conditions were either restrictive of the movement of the fallout, or the fallout was directed away from the Australian mainland (usually north west of Montebello Islands into the Indian Ocean).

However, while many of the ‘clouds’ moved out into the Indian Ocean, some moved across continental Australia. Following the detonation of the first Mosaic test the ‘cloud’ moved eastwards along the latitude of 20°S. Subsequently, there were later indications that it had passed over New Zealand (Butement et al., 1957).

Background testing of sites used in the collection of fallout were also undertaken around the time of weapons testing. In 1957 samples were collected prior to the Antler tests, these showed levels of 19 mCi/km². This radiation was later attributed to the Buffalo trials (Kearney et al., 1958).

No detail on the plutonium ratios in the weapons that were tested or in their fallout exists within the public domain, although it is known that the Totem and Mosaic had different proportions of ²⁴⁰Pu (Child and Hotchkis, 2013).

Since the deposition of caesium-137 from these events nearly two half-lives have passed. This makes it increasingly difficult to detect caesium accurately. To combat this plutonium which has a similar depositional history and migration features is tested. This presents an alternative to measuring caesium (Child and Hotchkis, 2013). Furthermore, the additional isotopic ratio information provided by measuring plutonium isotope adds another layer of information regarding the source of the material.

1.2 Caesium and Plutonium fallout

The ratio between the plutonium-240 and plutonium-239 is representative of the origin of fallout. The average ratio for ^{240/239}Pu fallout is 0.176 (Kelley et al., 1999, Krey et al., 1976), with the results from Australian studies varying from this result (Child and Hotchkis, 2013, Tims et al., 2013). A recent continent wide study shows an average ^{240/239} Pu ratio of 0.135 for Australia (Tims et al., 2013). The Sydney Basin is yet to be specifically measured, resulting in the initiation of this project.

Global fallout ratios of plutonium isotopes have been estimated with values of ~0.18 for the ^{240/239} Pu. However, due to the large differences in nuclear operations, weapon tests and accidents local fallout values may differ significantly from the mean depending on local sources (Child and Hotchkis, 2013).

The different ratios are a result of the fission time in which the plutonium-239 has been exposed. For weapons this is a short time (seconds) resulting in only a small amount of neutron capture and the creation of small amounts of plutonium-240. This is why low ratios occur around test sites. Higher ratios are the result of longer fission times such as those that would be expected at nuclear reactors where the plutonium may be irradiated for a number of years. Caesium-137, like plutonium-240 is the result of the fission and neutron capture that occurs within an atomic weapon and ratios of caesium-137 to plutonium-239 are also used. Though this study will focus on the more commonly used ^{240/239} Pu ratio.

Table 1.2 shows values taken from Kelley et al (1999) indicating the variations in plutonium isotope ratios as a function of latitude. This suggests that Sydney (and much of Australia) should have ^{240/239} Pu ratios of 0.185, higher than the mean. Kelley’s global study also found

Brisbane and Melbourne to have ratios of 0.1768 and 0.1716, which does not fit with an average of 0.185 representing southern regions. This and other studies of late have found that the values for plutonium isotope ratios in Australia are much lower than the global mean. This may indicate that Australian soils contain not global fallout but that of a localised atomic testing regime (Child and Hotchkis, 2013).

Table 1.2: Regional average compositions of Pu isotopes in soil (Kelley et al., 1999).

| REGION | LATITUDE | ATOMIC RATIO ^{240/239} Pu |
|------------------------|----------|------------------------------------|
| NORTHERN | 71-30 N | 0.180 ± 0.014 |
| NORTHERN EQUATORIAL | 30-0 N | 0.178 ± 0.019 |
| SOUTHERN EQUATORIAL | 0-30 S | 0.173 ± 0.027 |
| SOUTHERN | 30- 53 S | 0.185 ± 0.047 |

Since Kelley's (1999) article newer values have been reported presenting values for Australia that are much lower than the global mean, with average ratios for Brisbane reported as 0.15. Furthermore all the results of the continental study conducted by Tims et al. (2013) have found that the values for Australia are significantly lower than that of the global mean (Tims et al., 2013).

Child and Hotchkis (2013) have measured the plutonium levels and isotope ratios for areas surrounding the weapons testing in Australia from the 1950's- 60's using Accelerator Mass Spectrometry (AMS). By using samples from the archives of the Australian Radiation Protection and Nuclear Safety Agency, along with new samples they have been able to assess the regional impact of the testing. It was found that the fallout from the Totem and Mosaic testing are distinctly different from the global mean ^{240/239} Pu isotope values of 0.05 for Mosaic and 0.023 for Totem test sites. These will therefore influence the local fallout ratios (Child and Hotchkis, 2013).

While a global average of around 0.18 is accepted, different sources may differ significantly. For example, the composition of the plutonium from the accident at the Chernobyl reactor in the Ukraine has ratios ranging from 0.186 to 0.348. Levels above the mean are also seen in the ratios of the North Pacific Proving Grounds which measured ratios of 0.199 -0.22 (Child and Hotchkis, 2013).

By measuring the Plutonium isotopic ratio at a fallout site along with the isotopic ratio of the sources, it is possible to determine the source from which the fallout originated using the following formula (Krey et al., 1976).

$$\frac{(\text{plutonium activity})_1}{(\text{plutonium activity})_2} = \frac{R_2 - R}{R - R_1} * \frac{(1 + 3.60R_1)}{(1 + 3.60R_2)}$$

Where R= the ^{240/239} Pu ratio at the site and R1 and R2 are the ratios at the sources. As the sources are currently undefined, this will not be implemented for this project.

1.3 Chemical behaviour and analysis

^{137}Cs adsorbed on soil is strongly bonded and the amount of caesium in soils will only dramatically change as the result of natural decay or physical soil movement (Zhao et al., 2012, Ashraf et al., 2014). In soils Caesium-137 can be effectively adsorbed by clay minerals, particularly illite (Ashraf et al., 2014). However, this adsorption can be diminished by the presence of organic matter, thus suggesting that caesium is more mobile in soils with high organic contents (Łokas et al., 2013). It has been found that ^{137}Cs is least mobile in soils that are low in organic content and of a neutral to acidic pH, and in the presence of micaceous minerals containing free carbonates (Loughran et al., 1990).

Similar to caesium, plutonium is relatively immobile in soil, although in different chemical forms. There have been correlations found between the amounts of organic content and the concentration of plutonium found in soils. Soils are found to have higher plutonium concentration if there is more organic content in the soil. As fallout radionuclides are deposited on the soil surface, the levels of plutonium and other radionuclides will also depend on depth, often with an exponential decline occurring after 10cm (Tims et al., 2013). The amounts of radioactive caesium-137 deposited were originally nearly two orders of magnitude higher than the plutonium, and as a result caesium-137 has typically been measured using gamma spectrometry. Due to the mode of radioactive decay of plutonium this is not possible, and so methods that involve counting the atoms (e.g. AMS) rather than the decay are favourable (Everett et al., 2008), although alpha spectrometry can be routinely used for samples with sufficiently high plutonium content.

Passive Gamma Ray Spectrometers can be used in the measurement of erosion and deposition of soils. They are able to detect the distribution of the radionuclides such as ^{137}Cs that act as tracers in erosional studies (Stockmann et al., 2012). Gamma ray spectrometry works on the principle that photons have a discrete energy that is characteristic of its source isotope. The abundance of caesium reflects the geochemical variations in the surface layer of soils. Typically, mapping using gamma ray spectrometry is limited to potassium, uranium and thorium, but it can also be extended to the man-made radionuclide of ^{137}Cs by focusing on the energy peak of 662keV (Stockmann et al., 2012, Łokas et al., 2013). The measurement of isotope ratios in plutonium samples requires analysis by high sensitivity mass spectrometry techniques such as Accelerator Mass Spectrometry (AMS) (Child and Hotchkis, 2013).

During the measurement of radioactive elements radioactivity is generally given per dry mass. To achieve this, samples must be dried at 105°C until constant mass is achieved. Organic contents can be determined by Loss on Ignition (LOI) testing where the sample is heated to 600°C. As with most chemical analysis quality of the testing can be assured by the testing of both standard samples and blanks with the same process (Łokas et al., 2013). Prior to analysis solid samples are ground to ensure a homogenous mixture. Samples are also heated in order to eliminate organic fractions (Child and Hotchkis, 2013).

Following Maxwell's (Maxwell, 2006) methods to determine actinides in water Harrison (et al. 2010) has been able to measure radionuclides such as plutonium in a way that is suitable for an environmental matrix. Chemicals are separated using packed resin cartridges connected in series. Following digestion of the soil matrix with acid and precipitation Maxwell (2008) uses a series of columns each containing 2mL of TEVA, TRU, and DGA Resin. The cartridges were stacked on the vacuum box in the order given (Maxwell, 2008). Following the digestion stage Harrison et al. (2011) concentrates the plutonium using a calcium phosphate precipitation. This method resulted in detection limits of <0.075 Bq/kg for solid samples and much lower for liquids (Harrison et al., 2011).

1.4 Similar national and international case studies

There are few similar studies available for comparison to the current study. Below are some national and international studies that have looked into possible baseline levels of plutonium for a number of reasons. However, these are generally sampled from sites separated by long distances and representing different geographical regions. Soil and silt samples taken in the 1950's following the Mosaic testing where unable to find any increase in radioactivity in the catchments of the East Coast beyond that of the background radiation (Butement et al., 1957). These results were most likely limited by the technology of the time, especially as they were only able to measure total radioactivity, and not specific radionuclides.

This study is very similar to and partly modelled on a study that was conducted by Tims et al. (2013) of the plutonium levels in soils across continental Australia. This involved collecting samples from 14 distinct and undisturbed areas spread across the Australian mainland covering a range of climates, soil types and catchment sizes. This study involved 14 study sites though each of these represent 10 soil samples (Tims et al., 2013). Tims et al. (2013) collected soil from 0-2cm in depth from approximately 10 locations within each site with up to a few kilometres apart. It was found that plutonium levels varied very little with depth of soil, so taking samples from near the surface was sufficient to measure the whole fallout.

Internationally a study on peat in the Arctic reported a decline after 10 cm for the major radionuclides of americium, caesium and plutonium (Łokas et al., 2013). Similarly to soil, peat deposits are potentially important archives for anthropogenic radioactivity as mosses and peats effectively capture and retain airborne deposition. In this study conducted in the High Arctic, caesium-137 and plutonium ($^{239+240}\text{Pu}$ and ^{238}Pu) were recorded from peat measuring at 1.5 kBq/m², 13.3 kBq/m² and 0.3 kBq/m². These readings are linked to the atomic testing in the twentieth century along with the Chernobyl accident (Łokas et al., 2013). These are much higher than the plutonium and caesium which have been measured in Australia.

In the Sydney region studies have been conducted on the areas surrounding the HIFAR reactor at Lucas Heights, Sydney. These are predominately to study the impacts of a legacy waste site associated with the former Australian Atomic Energy Commission. Here there has been the measurement of other radionuclides along with plutonium, with samples taken from both soil and water and analysed using alpha source radiation (Payne et al., 2013, Harrison et al., 2011). Recent studies on waste trenches at Lucas Heights (Sydney) have found that the

levels of Plutonium are higher at the ground surface. This indicates that plutonium is mobile and able to move to the surface (Payne et al., 2013). This plutonium is distinct from global fallout.

1.5 Application to scientific research

Global fallouts have been used extensively for geomorphological research (Everett et al., 2008, Hancock et al., 2011, Hoo et al., 2011, Loughran et al., 1990, Simms et al., 2008a, Simms et al., 2008b, Stockmann et al., 2012, Zhao et al., 2012). Fallout has enabled us to distinguish between soils and sediment layers that have formed before the advent of nuclear weapons and those that have formed since. Plutonium is increasingly popular for this type of work as it has a similar depositional history and migration rates as caesium, but with a much longer half-life (24110 years for ^{239}Pu and 6561 years for ^{240}Pu to ^{137}Cs 's 30.07 years). This longer half-life means that a greater proportion of plutonium remains from nuclear weapons than the caesium which has decayed almost two half-lives since the atomic testing of the 1950's, making its detection hard in comparison to the more long-lived plutonium (Child and Hotchkis, 2013). Plutonium measurements are currently being used to assess the balance between soil formation and loss by identifying the sources of sediment based on different land uses (Tims et al., 2010). Many areas of Australia are under intensive land use and soil loss has become a problem, with massive erosion rates tipping the balance against soil formation (Tims et al., 2013).

^{137}Cs has been efficient as a tracer due to its low mobility and tendency to adsorb to small soil particles. It has been found that while the caesium binds to the mineral fraction, plutonium binds strongly to the hydrous oxide coatings of soils making an even stronger bond (Tims et al., 2013). These elements are able to retain these bonds during transportation making them ideal for the assessment of erosion and sedimentation (Everett et al., 2008).

The use of ^{137}Cs measurements affords a valuable means of assembling spatially distributed information on medium-term (c.40 years) rates of soil erosion. The ^{137}Cs approach has been applied successfully in a wide range of environments in many different areas of the world (Ritchie and McHenry, 1990, Wallbrink and Murray, 1996). These studies indicated that soil loss rates can be assessed with high accuracy on the basis of a single site visit. The approach is based on the key assumption that the local fallout is uniformly distributed and that subsequent remobilisation of ^{137}Cs reflects sediment movement (Simms et al., 2008a). Locally, studies have been conducted measuring the ^{137}Cs in catchment soils of the Sydney Basin. These studies have begun to create a caesium base level for the region that this project hopes to improve. However these numbers suggest that there may be a high variability in caesium within the environment. The values for caesium can be seen in the table below taken from Simms et al. (2008) and appear to average at around 5 Bq/kg ranging from 1 to 17 Bq/kg (Table 1.3) for a catchment to the south of the Sydney Basin.

Table 1.3 Caesium-137 activities for the Cordeaux Catchment, south of Sydney (Simms, Woodroffe et al. 2008).

| Sample | soil depth | ¹³⁷ Cs activity (BQ/kg) | error ± |
|--|------------|------------------------------------|---------|
| Reference ¹³⁷ Cs _{ref} | 10 | 9 | 1 |
| Kembla creek | | | |
| MB1 | 10 | 8 | 1 |
| MB2 | 10 | 10 | 1 |
| MB3 | 10 | 11 | 1 |
| MB4 | 9 | 9 | 1 |
| MB5 | 6 | 1 | 1 |
| MB6 | 9 | 10 | 1 |
| MB7 | 9 | 8 | 1 |
| MB8 | 8 | 8 | 1 |
| MB9 | 10 | 5 | 1 |
| MB10 | 9 | 4 | 1 |
| MB11 | 7 | 6 | 1 |
| MB12 | 6 | 5 | 1 |
| MB13 | 8 | 1 | 1 |
| MB14 | 10 | 5 | 1 |
| MB15 | 10 | 3 | 1 |
| MB16 | 10 | 8 | 1 |
| Kentish Creek | | | |
| KH1 | 10 | 3 | 1 |
| KH2 | 10 | 5 | 1 |
| KH3 | 10 | 6 | 1 |
| KH4 | 8 | 4 | 1 |
| KH5 | 9 | 9 | 1 |
| KH6 | 9 | 6 | 1 |
| KH7 | 10 | 4 | 1 |
| KH8 | 10 | 4 | 1 |
| KH9 | 10 | 4 | 1 |
| KH10 | 10 | 6 | 1 |
| KH11 | 9 | 8 | 1 |
| KH12 | 8 | 5 | 1 |
| KH13 | 9 | 17 | 2 |
| KH14 | 9 | 5 | 1 |
| KH15 | 10 | 6 | 1 |
| KH16 | 10 | 4 | 1 |

While caesium-137 has commonly been used for these studies it requires a large sample volume of ~100g and gamma spectrometry counting times up to 2 days. For a comparative sensitivity plutonium-239 can be measured using only 4 g of soil and AMS counting times of several minutes. This makes plutonium increasingly popular over caesium-137 for erosional studies which require many samples(Tims et al., 2010).

1.6 Potential impacts of Caesium and Plutonium

Environmental radiation baseline levels have raised extensive concerns, especially the great potential health risks of long half-life radionuclides in soil in regions with high density of residents such as areas of Northern China (Zhao et al., 2012). The effects of high levels of radiation, such as those experienced by the people exposed to the Hiroshima blast and the Chernobyl accident are complex issues beyond the scope of this paper. However, these include increased incidence in leukaemia, thyroid cancer and lung cancer. There have also been indications suggesting that the incidence of cataracts in children has increased in the area surrounding Chernobyl (Goldman, 1997).

The baseline studies are currently topical in the wake of the Fukushima accident, particularly in the Oceania region that is yet to experience the effects.

1.7 Natural and artificial abundance of isotopes

While the isotopes that are to be investigated within this study largely occur as the result of atomic fission and neutron capture, the elements caesium and plutonium also have naturally occurring isotopes. The only stable and naturally occurring isotope of caesium is ^{133}Cs . In this form caesium is found in the minerals beryl, avogadrite, rhodizite and economically in the mineral pollucite (Butterman et al., 2004).

While plutonium-239 is most abundant as an atomic product it also occurs naturally. Over geological time periods uranium will decay to produce naturally occurring plutonium. This decay leads to concentrations of approximately 1pg/g of plutonium-239 in uranium ores (Dixon et al., 1997).

1.8 Soils of the Sydney Basin

The soils of the Sydney Basin are old, having received little impact from the extensive erosion of the late Cenozoic due to the protection of the surrounding sandstone and the basins low relief (CSIRO Division of Soils, 1983). In this area the parent materials are derived mainly from the Triassic shales. The soils have strongly differentiated texture and an acidic nature. The major soil features are lateritic podzolic soils and thick weathered zones on relict surfaces. Red podzolic (purple in Figure 1.1) soils form in the well-drained sites above the shale, with yellow podzolic (yellow in Figure 1.1) and to a lesser extent greyed podzolic soils forming on the poorly drained sites. Above the sandstone the soils are dominated by podzols and humus podzols as well as modern alluvial soils (CSIRO Division of Soils, 1983). Towards the Blue Mountains and in the Royal National Park the soils become siliceous sands (light green) with lithosols (green) dominating towards Katoomba and Blackheath. (NSW Office of Environment and Heritage 2014).

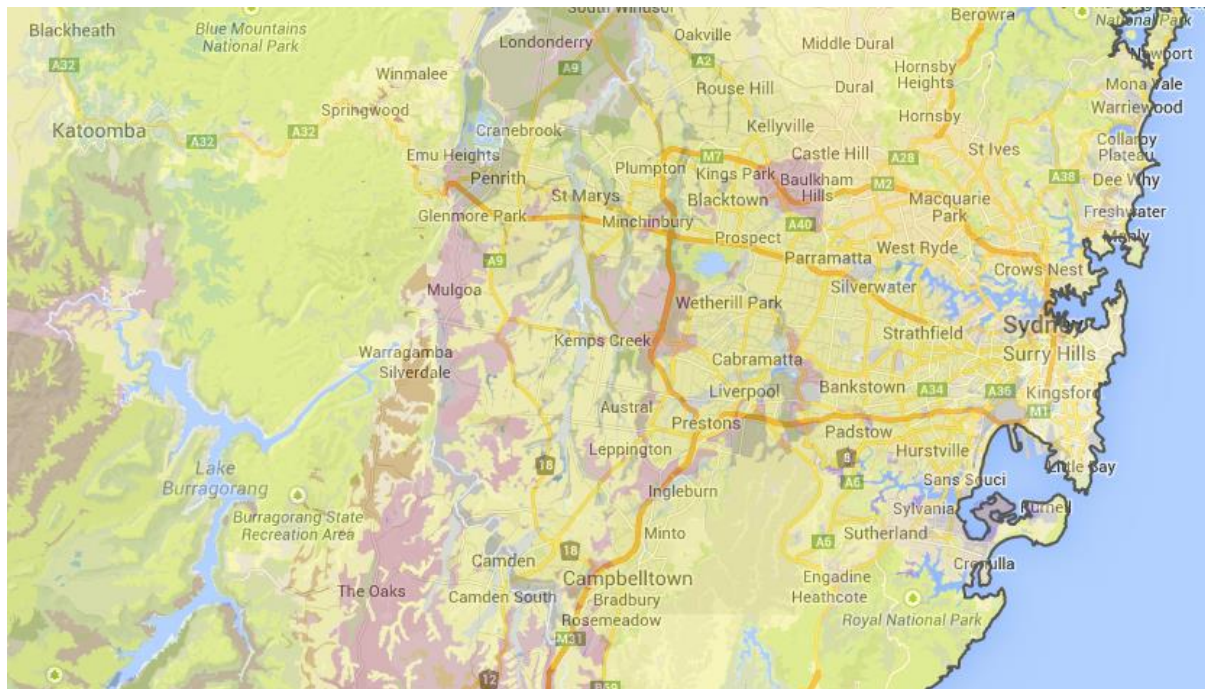


Figure 1.1 Soils of the Sydney Basin-Great soils classification (NSW Office of Environment and Heritage 2014) Red Podzolic soils are purple, Yellow Podzolic are in yellow, Siliceous Sands are in light green and the Lithosols are in the darker green.

Soils in some areas of the Sydney Basin have received significant contamination in the past. The soils around the inner city have been shown to have high levels of lead (Young and Young, 2001).

1.9 Summary of key points

Radionuclide fallout as the result of atomic testing in the 1950's and 1960's in Australia is known to have reached the east coast of the continent. It is important to generate a baseline for these data as a means of comparison both to other "unaffected" sites and to assess possible environmental impacts of past radioactive waste management and practices. The presence of fallout from atomic testing provides strong chronological markers in soils and sedimentation analysis. The ratios of the $^{240/239}\text{Pu}$ isotopes is representative of the source of the plutonium and in this study will be measured by Accelerator Mass Spectrometry (AMS). The global mean is ~ 0.18 with locations such as Chernobyl having higher than average ratios, while locations where weapons have been detonated having much lower values.

Chapter 2- Methods

A soil sampling regime was conducted across the Sydney Basin in order to determine baseline levels for activity concentrations and ratios of plutonium and caesium ($^{240/239}\text{Pu}$, $^{240+239}\text{Pu}$ and ^{137}Cs). Currently limited data on plutonium and caesium values in Australian soils exist, and these are sparsely spread across the continent (Child and Hotchkis, 2013, Tims et al., 2013), with only 2 values for NSW. In order to establish baseline levels for the Sydney Basin several sites were chosen, including representative samples of the entire basin, in terms of climate, lithology and topography. For this reason, an east west transect was chosen as this covered a large variety of local environments. The transect starts in the Royal National Park ($34^{\circ}04'56''\text{S}$ $151^{\circ}01'34''\text{E}$) and then heads west through Lucas Heights Science and Technology Centre (LHSTC, otherwise known as ANSTO) and finishing to the north-west in the Blue Mountains ($33^{\circ}38'52''\text{S}$ $150^{\circ}19'39''\text{E}$) (Figure 2.1).

Sampling of sites was conducted with the assistance of Professor Henk Heijnis, Dr Mathew Johansen and Ms Atun Zawadzki on the 21st and 26th of May 2014 and by Mr Jack Goralewski and Mr Sangeeth Thiruvoth on the 2nd of June 2014.

2.1 Regional setting

Sydney is located on the central east coast of New South Wales, Australia, and is home to approximately 4.8 million people. The region includes Australia's only nuclear reactor, the OPAL reactor that commenced operation in 2007, replacing the HIFAR reactor that had been in operation since 1958. Associated with this site is the Little Forest Legacy Site (LFLS) where low-level radioactive waste generated by the former Atomic Energy Commission was disposed of in trenches during a period between 1960 and 1968. To the west the region is bordered by the Blue Mountains which are over 1200m high while to the east the land forms cliffs that drop into the Tasman Sea.

2.2 Lithology and soils

The geology of the Sydney Basin is largely sedimentary dominated by the Triassic Hawkesbury Sandstone and Ashfield Shale. As caesium and plutonium have shown an affinity to smaller soil particles the study will focus on soil locations above the Ashfield Shale complex and the shales lenses in the Hawkesbury sandstone. These are both areas of lower relief and with finer soils. Clay soils such as those found at the LFLS offer a relatively favourable ion-exchange and sorption properties that appear to inhibit the migration of many radionuclides (Payne et al., 2013). Several samples for the present study were chosen to represent a similar lithology that will produce the clay soils (shales).

The nature of these areas is to produce fertile soils, and as such the majority of these areas have been cleared for agriculture, grazing and more recently housing (Young and Young, 2001). As a result of this the current study will also include areas where the base lithology is the Hawkesbury Sandstone.

2.3 Sampling Sites

Ideal sites are those which are pristine and therefore most likely to retain the entire fallout record accurately. Where possible sites were selected to represent areas that had had minimal disturbance since the 1950's when weapons testing was conducted both in Australia and around the world (Figure 2.1). Sites in national parks were preferred as they represent natural vegetation with minimal disturbance from humans. Additionally, a catchment was chosen adjacent to the LFLS where both soil and sediment were collected in order to establish a background site for the Lucas Heights location, in the vicinity of the LFLS



Figure 2.1 Sites selected for this study.

2.4 Sampling method

Samples were collected from the 12 sites representing the top 5cm of the soil. In order to collect representative samples of the possibly heterogeneous fallout radionuclides, samples were collected from three locations within a twenty metre radius of the site centre. Each of these samples consisted of four samples collected from within one square metre. All the subsamples were placed into a single bag and mixed to create a homogenous sample representing the site. At the site where sediment was collected samples were taken at points of fine sediment and kept separate for analysis. An algae sample was also collected along with the sediment sample from the Little Forest Background Creek.



Figure 2.2 Large tree used as a centre point of sampling, Woronora.

Samples were collected by placing a one metre square on the ground at a random distance (between 1-20 paces) from the central point. In order to collect the sample four locations within the square were cleared of leaf litter and an 80mm PVC pipe was hammered into the ground to 5cm depth. The soil around the pipe was removed with a metal spatula and then the sample was placed into the bag (Figure 2.3).



Figure 2.3 Sampling at Kings Tableland 26/5/2014. Present in photograph Henk Heijnis and Atun Zawadzki (holding PVC pipe used for sampling).

At each site a reference core has been taken in case of further study. In the event that uptake studies will also be conducted at another time, vegetation samples of small grasses are also collected at each site.

2.5 Sampling locations

Forest Brook

The Forest Brook site is located on the western edge of the Royal National Park bordering Heathcote (0317359E 6226422N UTM), forming the most easterly point of the transect. Sites further to the east have either been developed or have sandy soils. The site is located in an area of lush forest with a dense understory and almost closed canopy. The site was close to Forest Brook which is the head of a creek draining into the Port Hacking River. The site is on the border of the Heathcote shale lens and the Hawkesbury Sandstone and was chosen as this was the most pristine area that remained on the shale. The majority of the shale lenses in the area are covered by housing or highly disturbed parks and reserves. The area has a mean annual rainfall of 1102.2mm (Audley Bureau of Meteorology (BOM) 2014) and an elevation of 212m (Australian Height Datum (AHD)).

Forbes Creek Reserve

Located in Engadine (0317107 E 6230736 N) the Forbes Creek Reserve follows a steep gully that drains into the Woronora River via Loftus Creek. The area is partially situated on the shale lenses, though much of these have been eroded leaving the majority of the reserve on a sandstone lithology. The site is bordered by suburban housing on all sides and likely only remains due to it being too steep to build upon. Its closest BOM weather station is located at Lucas Heights and records a mean annual rainfall of 1010mm/yr. The site has an elevation of 109m (AHD).

Mill Creek

Mill Creek is a small creek located near the Little Forest Legacy Site (LFLS) at Lucas Heights. This creek drains from the Lucas Heights municipal landfill areas and joins Barden Creek which drains the LFLS and the northern side of the ANSTO site, eventually flowing into the Georges River. This sample was collected upstream of the Barden Creek confluence (0312588 E 6232859 N UTM), therefore the major anthropogenic influence on the creek is the municipal landfill. The geology of the area surrounding the creek is the Hawkesbury Sandstone. The closest BOM weather station is located at Lucas Heights (ANSTO) and the area has a mean annual rainfall of 1010mm/yr. The elevation of the site is 101m (AHD).

Little Forest Background Sites

As noted above, during the 1960's waste including radionuclides was buried in trenches in what is now known as the Little Forest Legacy Site (Payne, 2012). In order to establish a background comparison for this site, four samples were taken from an adjacent catchment area that does not drain the LFLS.

These involved one soil sample (0312482E 6232478N UTM) and three sediment samples from a small ephemeral creek which was dry at the time of sampling (0312706E 6232457N UTM). One of these sediment samples also featured dried algae that was also analysed. The soil site and the creek are located on the Hawkesbury Sandstone. The site has an elevation of 123m (AHD) for the soil sample and 121m for the sediment samples. The closest BOM weather station is located at Lucas Heights (ANSTO) and has a mean annual rainfall of 1010mm/yr. These sites appeared undisturbed at the time of sampling, but later investigation

has found that the surrounding areas (but possibly not the actual site of the soil sample) had been cleared extensively during the 1980's as shown by the aerial photography in Appendix 2.

Sandy Point

This site is located within a reserve in the Sandy Point area (0314646E 6238070N UTM). The site is situated above the Georges River with an elevation of 30m (AHD). The area has highly sandy soils as a result of the underlying Hawkesbury Sandstone, with places where there is no soil present at all. The closest BOM weather station is located at Holsworthy Control Range measuring a mean annual rainfall of 706.5mm/yr.

Woronora

This site is located in a bush area of the Prince Edward Park (0319292E 6232636N UTM), in the high ground behind the Woronora RSL. The site featured many large mature trees and was on relatively steep ground with flat sections. The sample was collected from one of these flat sections. The site has an elevation of 31m (AHD). The nearest BOM weather station is located at Lucas Heights (ANSTO) measuring a mean annual rainfall of 1010mm/yr.

Rossmore Grange

This site is located in the middle of Western Sydney (0292990E 6243742N UTM) and is the only point on the transect that occurs between the Georges River and the Nepean River. In this area undisturbed land is rare, as the underlying lithology is no longer the Hawkesbury Sandstone but the Ashfield shale. Here the soils are much more developed than their coastal counterparts. The Rossmore Grange site has in the past been cleared, but agriculture was abandoned before the 1950's. Later it was to be reclaimed by the Liverpool City Council as parklands. The site is home to remnants of Cumberland Plain vegetation (Smith et al., 2006) and the soil samples were collected from within this area to reduce the risk of disturbance. The closest BOM weather station is located at Badgerys Creek AWS and records a mean annual rainfall of 675.5 mm/yr. making it the driest location sampled. The elevation of the area is 60 m (AHD).

Bents Basin

As part of the Bents Basin Conservation Area (0281377E 6243150N UTM) this site was free from clearing, although the sample site may have formed part of a flood terrace and so the origin of the soil may be complex. Any disturbance to the land has probably resulted from natural causes. While this may have been an ancient flood terrace the trees which have been used as guides to the ages of the vegetation were very large, indicating it had been some time since the area had been disturbed. The site is situated above the Hawkesbury Sandstone very close to the Lapstone Monocline. The site has an elevation of 62 m (AHD) with the closest BOM weather station located at Wallacia Post Office, recording a mean annual rainfall of 884.6 mm/yr.

Glenbrook

The Glenbrook site (027875E 6258498N UTM) is located in the lower Blue Mountains. The site is located in an area of open forest with little to no understory present. The site was located well within the National Park and as such is unlikely to have been disturbed. There was however, evidence of fire with some of the larger trees being charred. The site has an elevation of 177 m (AHD). The closest BOM weather station is located at Glenbrook Bowling Club and records a mean annual rainfall of 981.5 mm/yr.

Kings Tableland

This site (0256970E 6262472N UTM) is located within the Blue Mountains National Park near the Queen Victoria Hospital. The site is located in a low open woodland/scrub with coarse sandy soils. These poor soils have resulted from the underlying lithology of the Narrabeen Group Sandstone. The elevation of the area is 873 m (AHD) and is the second highest measured in this sample series. The closest BOM weather station is located at Katoomba and records a mean annual rainfall of 1404 mm/yr., the highest of any of the sampled sites.

Evans Lookout

This site is located at the highest (elevation 1031m (AHD)) and most westerly point of the sampling transect (0251060E 6273194N UTM). The site is a ridge that drops steeply to both sides and is home to a closed woodland type vegetation. The site is located on the Narrabeen Group Sandstone though features better developed soils and vegetation than the Kings Tableland site. The nearest BOM weather station is located at Mount Boyce and records a mean annual rainfall of 961.1mm/yr.

2.6 Site assessments

Vegetation was assessed because it influences the deposition of radionuclides as well as pedogenic processes. Vegetation is assessed on structure, canopy cover and type, using tables of estimated crown coverage and structural formations for Australian vegetation, following Specht (Specht et al., 1974) . These tables used the height of the tallest species and the percent of canopy cover to determine a structural class. Large trees were used to assess the potential for disturbance (where the presence of large trees implied no disturbance for at least the trees lifetime) resulting in most of the areas falling into open forest and low open forest categories (areas with tall trees and a moderately open canopy). No grasslands were sampled due to the lack of reference to disturbance present in the vegetation. Canopy cover was not recorded for the sediment samples as it was not thought to be an indicator of disturbance for these sites, due to the sediment originating elsewhere.

Alluvial sediments represent a collection of eroded regolith from the surrounding catchment, and therefore potentially concentrates or dilutes atmospheric fallout from a catchment.

Descriptions of the specific locations of samples sites are taken to assess if the geomorphology of the landscape impacts on the amount of fallout that is present (see site descriptions). This is biased by the site selection that is often on high flat ground as this was deemed less disturbed by erosion and other factors. Rainfall plays an important part in the deposition of fallout so climate characteristics are gathered from other parties for each

location. Elevation of each site was assessed with the GPS (Explorist 110) as only general comparative elevations were required. The lithology of the sites crosses over from the Hawkesbury Sandstone to the shales of the Adaminaby Group before returning to the sandstone capping of the Blue Mountains.

2.7 Analysis

Several methods have been utilised in this study. The caesium-137 was measured using Gamma Spectrometry, plutonium isotopes (^{239}Pu and ^{240}Pu) by Accelerator Mass Spectrometry and combined $^{240+239}\text{Pu}$ by Alpha Spectrometry. To supplement this several methods were used to assess the soil properties of the samples including Loss on Ignition (LOI) for organic content, grain size analysis and pH.

As the literature (Tims et al., 2013, Everett et al., 2008) suggests that both Cs and Pu are more abundant in the finer fraction of the sample, samples were sieved to 63 microns in order to have the highest chance of detection.

2.8 Physical processing

Following collection, samples were sieved with a 2mm mesh sieve and run through a riffle. This unbiasedly divides the sample into halves. One of these is used for wet sieving and the other is retained in case of reanalysis. Samples were wet sieved with a 63 micron mesh sieve in order to concentrate smaller particles with which radionuclides are associated, allowing for greater likelihood of detection.

An initial test showed that the 63 micron sieve was too fine to be used alone (due to clogging) and it was faster to sieve in series. The wet sieving process involved the passing of the sample and around 2L of deionised water through a series of stacked sieves (250 microns, 125 microns, 63 microns). To ensure that the finer particles were not trapped within the coarser matrix this is done as a two 1L washes retaining the filtrate. The components that remained in the sieve and in the filtrate were kept separate and dried at 60°C for at least three days, then at 100°C for 24 hours to ensure complete dryness before being weighed.

Following drying the <63 micron samples formed solid cakes. In order to achieve best results with the gamma spectrometry and the AMS the less than 63 micron samples were ground into a fine powder using a mortar and pestle. This <63 micron fraction was used for further analysis. The > 63 micron fraction was weighed and retained without further analysis.

2.9 Gamma spectrometry

Processing for the gamma spectrometry is straightforward, as the sample does not require chemical pre-treatment (unlike AMS and alpha spectrometry). Therefore only the physical preparation and packing of samples was required. Dried and ground samples are packed into weighed petri dishes (65mm diameter or 35mm diameter according to the amount of sample available). These petri dishes were then sealed with a silica gel and allowed to equilibrate for three weeks. This seal and the equilibration time allowed for radon gases to equilibrate allowing for the measurement of U and Th series radionuclides as well as the ^{137}Cs from the same sample (although not reported in this present study, the U and Th data are useful additional information often obtained for soils).

Samples were analysed using a low energy gamma spectrometer, by a Compton Suppression Gamma Spectrometry System, for between one and three days. This measures the Gamma radiation emitted by the sample and used to calculate an activity concentration of the Caesium. The Canberra Compton suppression detector system comprises an active NaI(Tl) suppression annulus, a NaI(Tl) plug detector and a reverse electrode germanium (REGe) detector all housed within an inert lead shield.

^{137}Cs activity was determined using the 662 keV peak. Activities quoted are at the date of counting, with quoted uncertainties at 1σ counting errors and less than (<) values are quoted at the 95% confidence interval. The detector system efficiency calibration was carried out using a National Institute of Standards and Technology (NIST) traceable multi-nuclide standard source.

2.10 Accelerator Mass Spectrometry (AMS)

The source of plutonium in the environment can be determined by analysing the ratio of the 240 and 239 isotopes. Accelerator mass spectrometry (AMS) is able to measure Pu in environmental materials with great sensitivity, with detection limits on ^{239}Pu and ^{240}Pu at the femtogram level. While other techniques can be used for the analysis of plutonium at these levels, methods such as Alpha Spectrometry are unable to differentiate between the ^{240}Pu and ^{239}Pu isotopes. The ANTARES AMS facility at ANSTO, Lucas Heights was used for the analysis of these isotopes. Because of the high sensitivity of the AMS instrument and the expected low concentrations of plutonium expected in the samples, sample preparation was conducted in a clean laboratory facility at ANSTO. A flowchart summary of the process can be seen in Appendix 1.

Sample preparation was conducted according to the standard procedure. Samples are first ashed at 800°C for 8 hours to remove any remaining organics (samples were analysed for LOI at 600°C prior to further ashing 800°C so mass change was negligible). To remove the soil matrix samples were refluxed for 12 hours in Aqua regia (1:3 ratio of Nitric and Hydrochloric acids) then centrifuged. The supernatant was taken and fumed down to near dryness on a hotplate three times with 2ml of concentrated nitric acid added at each drying. This was done in order to remove the chlorine that can impede retention of the Pu. The pH of the sample was raised to ~8 in order to form an iron hydroxide co-precipitate to concentrate the Pu prior to ion exchange purification. This precipitate was then washed with Milli-Q water 2-5 times to remove residual chlorine. The precipitates were then dissolved in 3M HNO_3 for loading onto ion exchange columns.

Gelatinous hydrated silica was found in the samples and was likely to cause the ion exchange resin to block. To remove the silica, 1mL of 0.5M polyethyleneglycol MW 2000 (PEG 2000) was added to each sample and samples were heated gently to dehydrate and polymerise the silica. The silica was then able to be removed via centrifuging.

Plutonium was extracted and purified from the samples using EiChrom TEVA® ion exchange resin. Prior to loading samples onto the columns, plutonium in all oxidation states (6+ to 4+) is reduced to 3+ using ferrous sulfamate. The plutonium is then oxidised back to

the 4+ state by addition of sodium nitrite, allowing the Pu to be strongly retained by the tetravalent specific resin. In this way the plutonium is able to be separated from other potentially interfering actinide species such as ^{238}U which exists as a poorly retained hexavalent species, and matrix components such as iron and calcium present as 3+ or 2+ ions. Uranium was captured in the eluent from the TEVA resin on a UTEVA® resin column. This will be analysed though not within the time frame of this study.

The columns were then washed with 3M HNO_3 , conditioned to chloride form with 9M HCl and washed with 5M HCl . Purified plutonium was then able to be eluted from the TEVA® column. Plutonium was collected using an Iron Hydroxide co-precipitate that was then dried in an oven at 60°C then heated in a furnace to convert the iron hydroxide precipitate to iron oxide (hematite). This was then mixed with niobium powder and packed into cathodes for AMS analysis. During the firing of one batch of samples the furnace malfunctioned resulting in the pellets of iron oxide becoming too hard to crush. As a result of this the uncrushed pellets were loaded into the cathode surrounded by niobium powder and pressed without mixing. This was not ideal but appears to have worked as output from these cathodes was equivalent to typical samples.

The samples were analysed by the ANTARES accelerator, the layout of which can be seen in Figure 2.4. As actinides do not readily form negative ions, molecular oxides are used in the ion source. These are analysed by a double focusing magnet which is equipped with an electrostatic isotope switching system (low energy bouncer). The beam is then injected into the FN Tandem accelerator (ANTARES) which is operated at 4MV for actinides with argon gas stripping located within the terminal. After acceleration the 5+ beam is selected and directed into the actinides beamline by the 12° electrostatic deflector. The beams are then analysed using a spherical-geometry electrostatic analyser for energy and a double focussing analysing magnet for mass. Isotopes are detected using a gas filled ionisation detector measuring each ions total energy (Hotchkis et al., 2010)

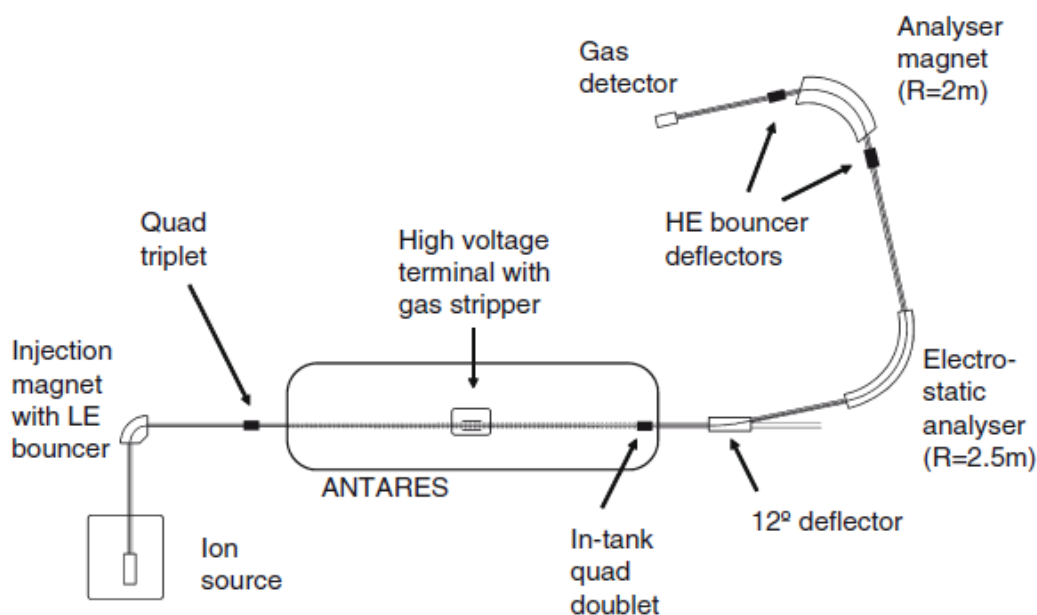


Figure 2.4 ANTARES accelerator used to analyse samples (Hotchkis et al., 2010).

Samples have also been run on the newer VEGA accelerator. This is similar to the ANTARES yet runs on 1MV instead of 4MV. This newer accelerator also measures the 3+ Pu stage, which gives better count rates than the 5+. There are several key differences between this accelerator and the ANTARES as this one is designed especially for heavy ion analysis. The VEGA accelerator has wider beam tubes to allow for the wider beam divergence associated with heavy ion analysis. There is also an additional electrostatic analyser on the low energy pre accelerator section. This reduces the scatter from the beam prior to acceleration. There is also an extra analysing magnet on the high energy section (post accelerator) which reduces the scatter caused by the breakup of molecules inside the accelerator, and a terminal vacuum pump that also reduces the scatter. The VEGA accelerator uses helium as a stripper gas in the accelerator terminal as opposed to the ANTARES which used argon or sulphur hexafluoride. The helium allows for greater efficiency in electron stripping.

2.11 Alpha Spectrometry

In order to compare the results achieved from the AMS analysis of the <63 micron sieved portion to the <2mm bulk samples (all the $^{239,240}\text{Pu}$ is in the <63 micron fraction, a set of 8 bulk samples were analysed for $^{239+240}\text{Pu}$ using alpha spectrometry. Samples were selected based on plutonium concentrations from the AMS results, factoring in the detection limits of the alpha spectrometry ($\sim 0.04\text{ Bq/kg}$), these were then dried and ground finely before digestion and addition of tracers. Samples were prepared according to Harrison et al. 2011. Soil samples were digested using Aqua Regia, co-precipitated with $\text{Ca}_3(\text{PO}_4)_2$ and dissolved in 3M HNO_3 . The plutonium was separated once again using the TEVA® resin after fixing oxidation state of plutonium to Pu (+4). The Pu fraction, elute from the TEVA® resin was evaporated to near dryness, diluted with Milli-Q (18.2 MΩ) water. A cerium fluoride micro-precipitate alpha source was prepared by adding cerium carrier and hydrofluoric acid and collecting the resultant precipitate on a 0.1 μm filter paper.

Alpha sources were counted for 7 days in a Canberra Alpha Analyst™ fitted with Passivated Implanted Planar Silicon (PIPS) detectors. Energy and efficiency calibrations for each alpha detector were performed using a NIST traceable multi alpha standard mixed stainless steel source (^{238}U , ^{234}U , ^{239}Pu and ^{241}Am ; stainless steel source 7400-SRC).

2.12 Additional analysis

In addition to measuring radioactivity and concentration of the Pu and Cs, the collected samples were analysed for grainsize, pH, moisture and organic content as these factors may influence the Pu and Cs measurements.

Grainsize

Samples were sieved and measured in greater than 2 millimetres, less than 2 millimetres but greater than 63 microns and less than 63 micron components. From this it is possible to compare the dominant grain sizes of each of the samples.

In addition to sieving the grainsize of each sample was measured using a Malvern MasterSizer 2000 laser sizing instrument. In preparation for this the samples needed to be sieved to less than 1 millimetre as this was the limit of the machine. Samples were then reacted with H_2O_2 in order to remove the organic components. Following this 1mL of sodium phosphate (0.5M) was added to aid sample dispersal and the sample agitated in an ultrasonicator bath for 15 minutes. Subsamples were then analysed using the MasterSizer 2000.

pH

The pH of the <2mm sieved soil was measured using ISO10390:2005, which is the current international standard for the measurement of soil pH. This involved drying at 40°C, adding 5mL of dry soil to 25mL of Milli-Q (18.2 MΩ) water and shaking for 1 hour. The sample was then left to sit for another hour before the pH of the water was measured using a Metrohm AG 9101 Herisau glass electrode connected to a 781 pH/Ion meter console which calibrated with Metrohm buffers 4, 7 and 9 prior to use. The pH was tested three times and the average value taken. For the average to be acceptable the standard deviation of the three measurements must be below 0.2 pH units. All samples were below this threshold with the majority of samples having a standard deviation of less than 0.1.

Moisture

The moisture content of the samples was determined based on weight prior to sieving and then the residues were weighed after drying. This allows the moisture content to be determined. The smaller portion of the riffle was left undried in the event that the samples had to be reanalysed. Because of this the loss due to drying of the larger sample was taken as representative of the whole.

Loss on Ignition (LOI)

The samples were analysed for loss on ignition in order to identify the amount of organics present. To do this around 8-10g of sample was heated to 600°C in a furnace and left for 24 hours. The change in the mass of the sample as a result of this is counted as the Loss on Ignition and represents the removal of organic components of the sample.

2.13 Summary

14 samples have been collected from the Sydney Basin and Blue Mountains and have been analysed by Gamma spectrometry for caesium-137, AMS and alpha spectrometry for, plutonium-240 and plutonium-239 and AMS for the $^{240/239}$ Pu ratio. One sediment sample also included large amounts of dried algae that were also analysed.

Preference was given to undisturbed sites on shale ridge tops, though these sites were not always possible due to clearing. Because of this many sites were instead undisturbed sites on flat sandstone areas.

Table 2.1 Summary of selected sites.

| Sample | Sample type | Location type |
|--------------------------------------|------------------------|-----------------------|
| Forest Brook | Soil –Earthy sands | Flat ridge top |
| Woronora | Soil – undetermined | Flat – mid slope |
| Forbes Reserve Engadine- | Soil- Siliceous sands | Flat mid slope |
| Little Forest Background - | Soil - Yellow Podzolic | Flat ridge top |
| Little Forest Background- creek 1 | Alluvial Sediment | Dry creek |
| Little Forest Background- creek 2 | Alluvial Sediment | Dry creek |
| Little Forest Background- algae | algae | Dry creek |
| Little Forest Background- creek 3 | Alluvial Sediment | Dry creek |
| Upper Mill Creek | Alluvial Sediment | High section of creek |
| Sandy Point | Soil- Yellow Podzolic | Flat ridge top |
| Rossmore Grange | Soil- Yellow Podzolic | Flat flood plain |
| Bents Basin | Soil- Siliceous sands | Flat – river terrace |
| Glenbrook | Soil- Siliceous sands | Flat ridge top |
| Kings Tableland | Soil- Siliceous sands | Flat ridge top |
| Evans Lookout | Soil- Siliceous sands | Flat ridge top |

Chapter 3 -Results

Fourteen samples of either soil or alluvial sediment along with one dried algae were analysed for caesium-137 and plutonium-239+240 as well as the $^{240/239}$ Pu isotope ratios. In conjunction with these tests, environmental factors were measured to determine their impacts of the retention of fallout.

As the samples were sieved the data in the Table and Figures below is presented as Bq/kg for the dry <63 micron component. These values are not adjusted for the full soil mass (>63 microns) as it was shown through alpha testing of the plutonium for the bulk samples, that while the majority of the plutonium was contained within the <63micron fraction the value varied. The adjustment of these samples assuming that all the fallout was retained in the fine fraction could result in values that are much lower than reality and with no certainty.

3.1 Caesium

The highest caesium was recorded for the Forbes Creek Reserve site with 25.7 Bq/kg (± 1.1) (Figure 3.1). Generally the alluvial sediment samples show very low levels of caesium with values of less than 3.5 Bq/kg, although low levels can also be seen in the Bents Basin and Rossmore Grange sites. The lowest value recorded was for Rossmore Grange with 0.6 Bq/kg. Possible reasoning for the variation in values will be discussed later.

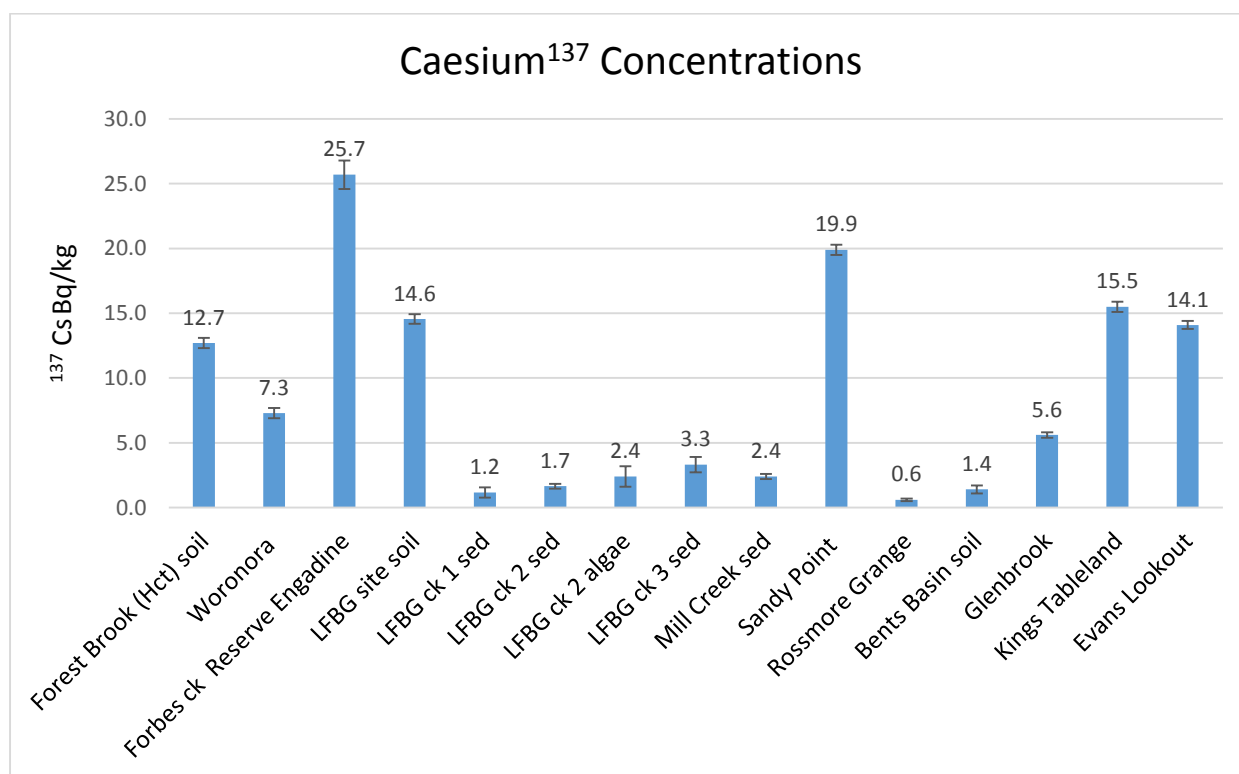


Figure 3.1 Caesium activity at sites sampled in this study. LFBG is an abbreviation of Little Forest Background.

3.2 Plutonium

The plutonium is present in much lower abundance than the caesium with the highest plutonium being recorded for the Kings Tableland and Evans Lookout samples with a value of 0.52 Bq/kg for each. The lowest soil value was recorded for the Bents Basin site with a result of 0.053 Bq/kg, the lowest sediment result was 0.02 Bq/kg (Figure 3.2). The average of all the sites is 0.24 Bq/kg (standard deviation 0.2). The average for the soils alone was 0.31 Bq/kg (standard deviation 0.19) and 0.05 Bq/kg (standard deviation 0.03) for the sediments. In order to test whether the plutonium was in fact concentrated within the <62 micron fraction a selection of samples with higher plutonium were also analysed by Alpha Spectrometry (Figure 3.3). This showed values for the $^{240+239}\text{Pu}$ ranging from 0.04-0.2 Bq/kg. These are much lower than those that were measured for the <63 micron fraction, indicating that there was concentration of the plutonium with the finer particles.

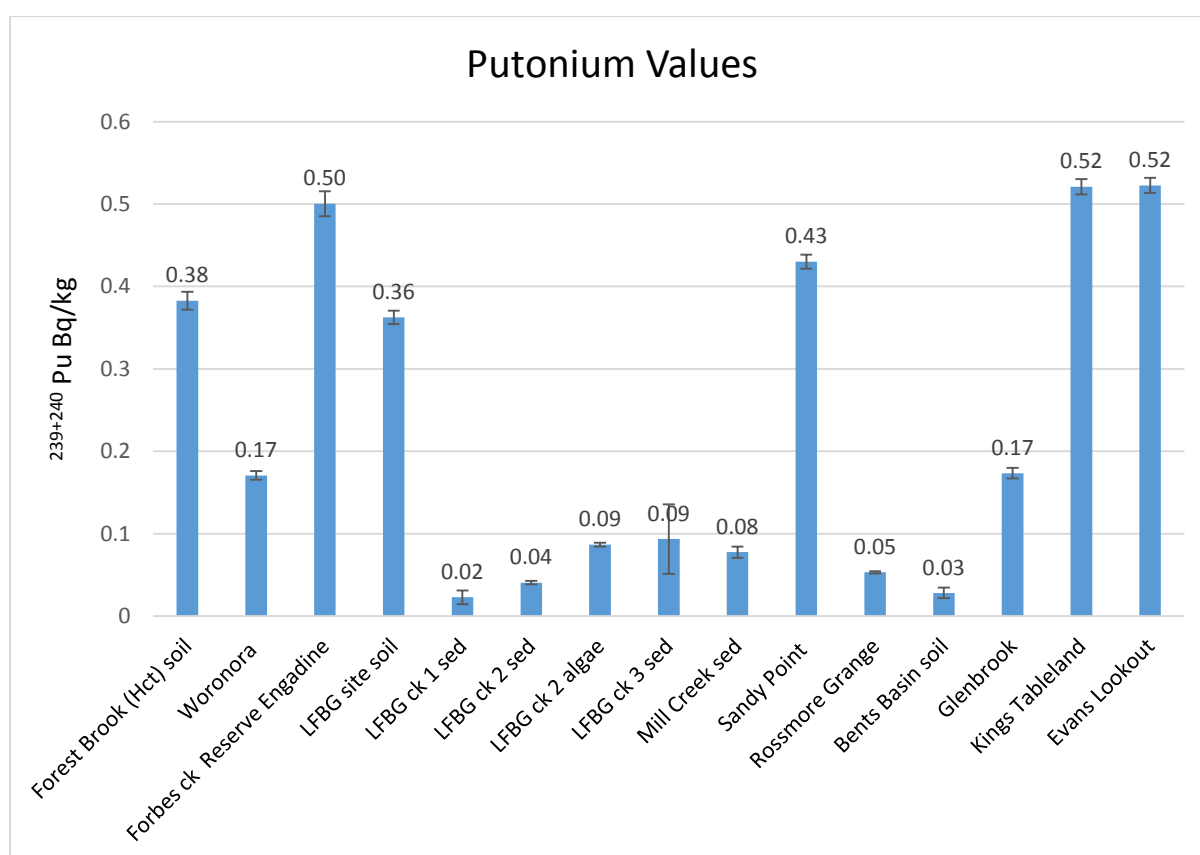


Figure 3.2 Plutonium activity at the sampling sites (<63Micron fraction).

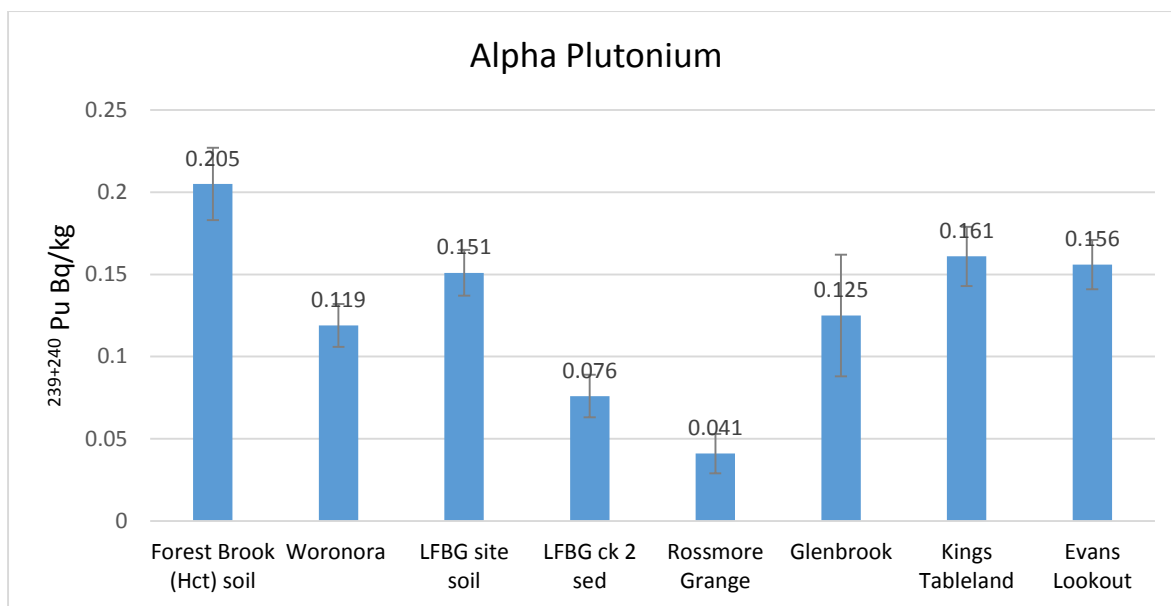


Figure 3.3 Bulk sample Plutonium assessed by Alpha spectrometry. Only these 5 samples were tested due to the AMS results indicating that the others would be below the detection limits of the Alpha analysis.

The relationship between the plutonium and the caesium is shown in Figure 3.4. It shows that there is a linear relationship between the two radionuclides. Confirming that they are interrelated with a fit of 87% ($r^2=0.867$). The values for both the Plutonium and the Caesium can be seen in Table 3.1.

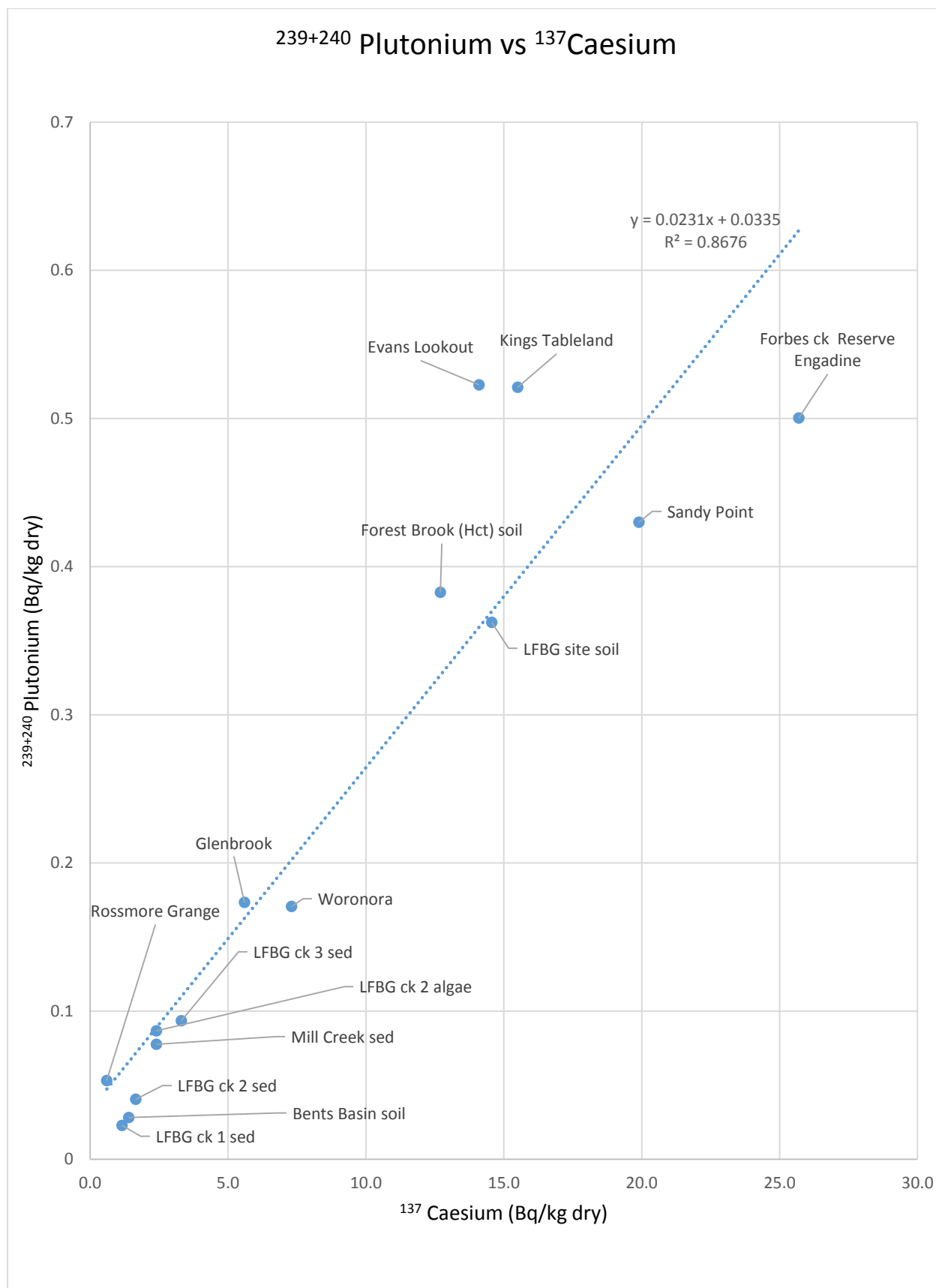


Figure 3.4: Relationship between the Caesium and the Plutonium activity measured for the samples.

Table 3.1: Caesium and Plutonium activity for each site. As the values are reported for the concentration within the <63microns fraction, the percentage of the total sample that this fraction represents is also included.

| Sample | Plutonium-239+240 | | Caesium-137 | | Fraction <63 microns |
|--|-------------------|-------|-------------|-------|-------------------------|
| | Bq/kg | Error | Bq/kg | Error | |
| Forest Brook-soil | 0.383 | 0.003 | 12.7 | 0.4 | 9% |
| Woronora -soil | 0.171 | 0.002 | 7.3 | 0.4 | 37% |
| Forbes Reserve Engadine-soil | 0.500 | 0.006 | 25.7 | 1.1 | 9% |
| Little Forest Background -soil | 0.362 | 0.008 | 14.6 | 0.4 | 23% |
| Little Forest Background-creek sediment 1 | 0.022 | 0.000 | 1.2 | 0.4 | 34% |
| Little Forest Background-creek sediment 2 | 0.040 | 0.000 | 1.7 | 0.2 | 35% |
| Little Forest Background-algae | 0.087 | 0.017 | 2.4 | 0.8 | |
| Little Forest Background-creek sediment 3 | 0.094 | 0.002 | 3.3 | 0.6 | 8% |
| Upper Mill Creek-sediment | 0.078 | 0.001 | 2.4 | 0.2 | 21% |
| Sandy Point –soil | 0.430 | 0.008 | 19.9 | 0.4 | 8% |
| Rossmore Grange | 0.053 | 0.001 | 0.6 | 0.1 | 45% |
| Bents Basin- soil | 0.028 | 0.001 | 1.4 | 0.3 | 17% |
| Glenbrook-soil | 0.173 | 0.002 | 5.6 | 0.2 | 45% |
| Kings Tableland -soil | 0.521 | 0.009 | 15.5 | 0.4 | 14% |
| Evans Lookout-soil | 0.522 | 0.009 | 14.1 | 0.3 | 14% |

3.3 Plutonium Isotopes

Initially only 8 of the analysed 15 samples (including the algae) were able to produce accurate $^{240/239}\text{Pu}$ isotope ratios. The others are believed to have suffered from both low count rates and contamination from uranium that falsely registers on the AMS detector inflating the ^{240}Pu value making the ratio inaccurate. Results from these contaminated values suggested that the soil contained pure reactor grade plutonium, which was extremely unlikely. These were rerun using the new VEGA accelerator at ANSTO. This achieved clearer results with much smaller errors. The exception to this was the algae sample, which failed to produce a reliable isotope ratio due to small sample mass. The isotope ratios range from 0.117 for Forest Brook to 0.162 for the Little Forest Background soil. This places them well below the global average of 0.176 (Figure 3.5). The values have been adjusted for LOI due to the pre-analysis processing requiring the samples to be ashed. This results in the output from the AMS as showing concentration per mass of ashed sample, as the caesium was not ashed LOI adjustments were made for the plutonium.

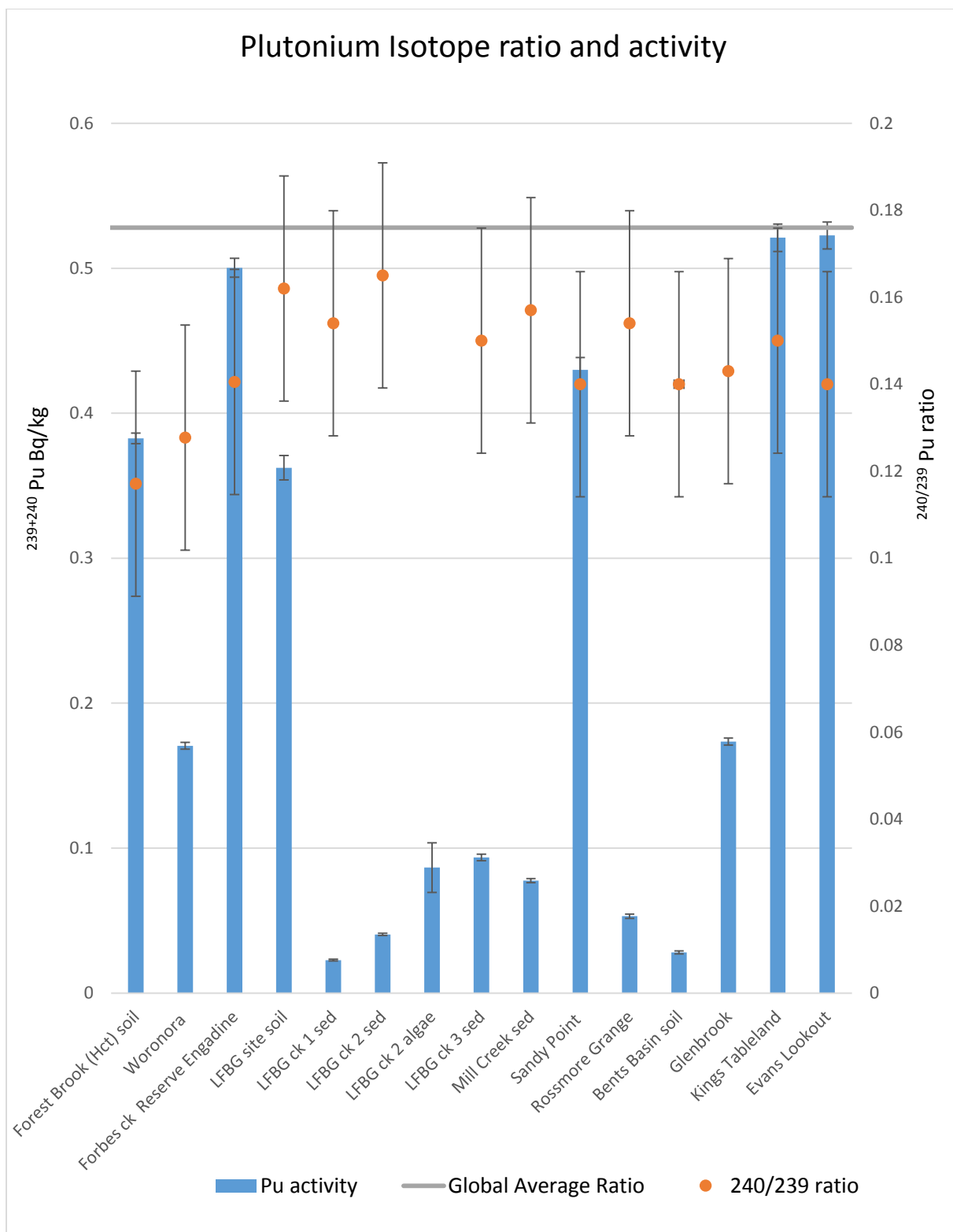


Figure 3.5: Pu concentrations and corresponding isotope ratios for the Sydney sites. Concentrations are for the <63 micron fraction of the soil and adjusted for LOI. Algae isotope ratio was inconclusive.

3.4 Moisture Content

It was found that the moisture content of the samples varied appreciably and showed no correlation to an east west trend. The Mill Creek sample was extremely wet with water visibly settling out of the sample between collection and analysis. This runoff was not included in the moisture analysis. Water content represented 8-38% of the overall sample mass collected (Figure 3.6). The sediments collected showed the same variation as the soil samples with the LFBG creek samples, showing a trend of drying downstream. While there appears to be no trend between the caesium-137 or plutonium-240+239 and moisture (Figure 3.7).

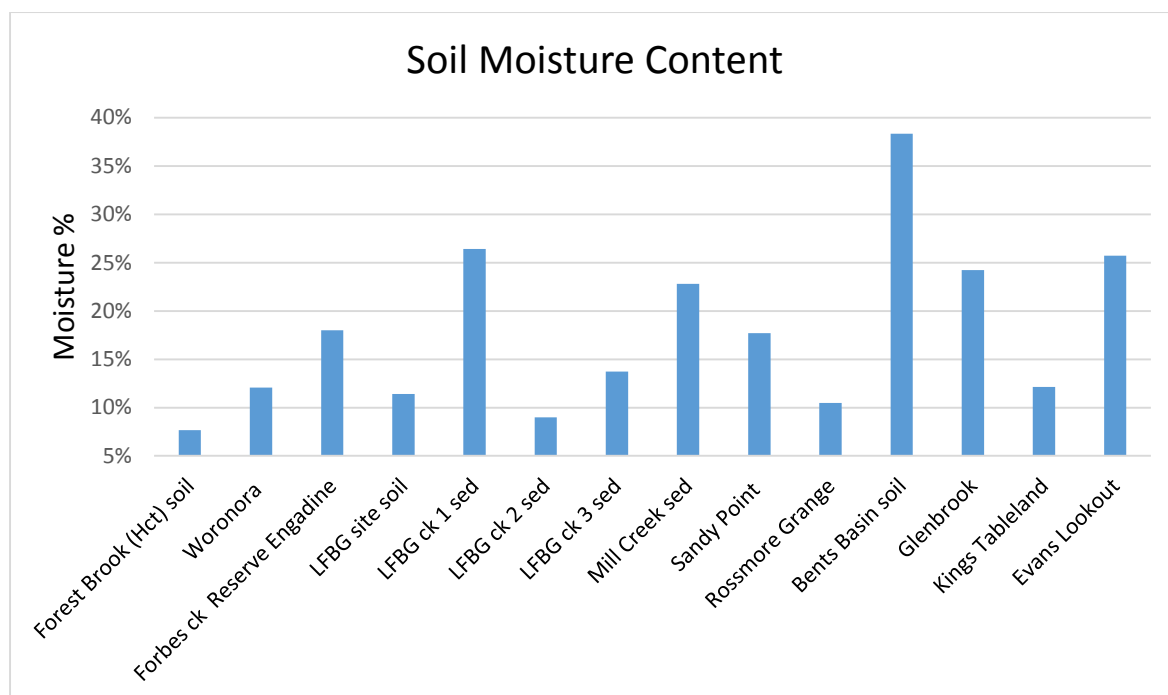


Figure 3.6: Soil moisture content.

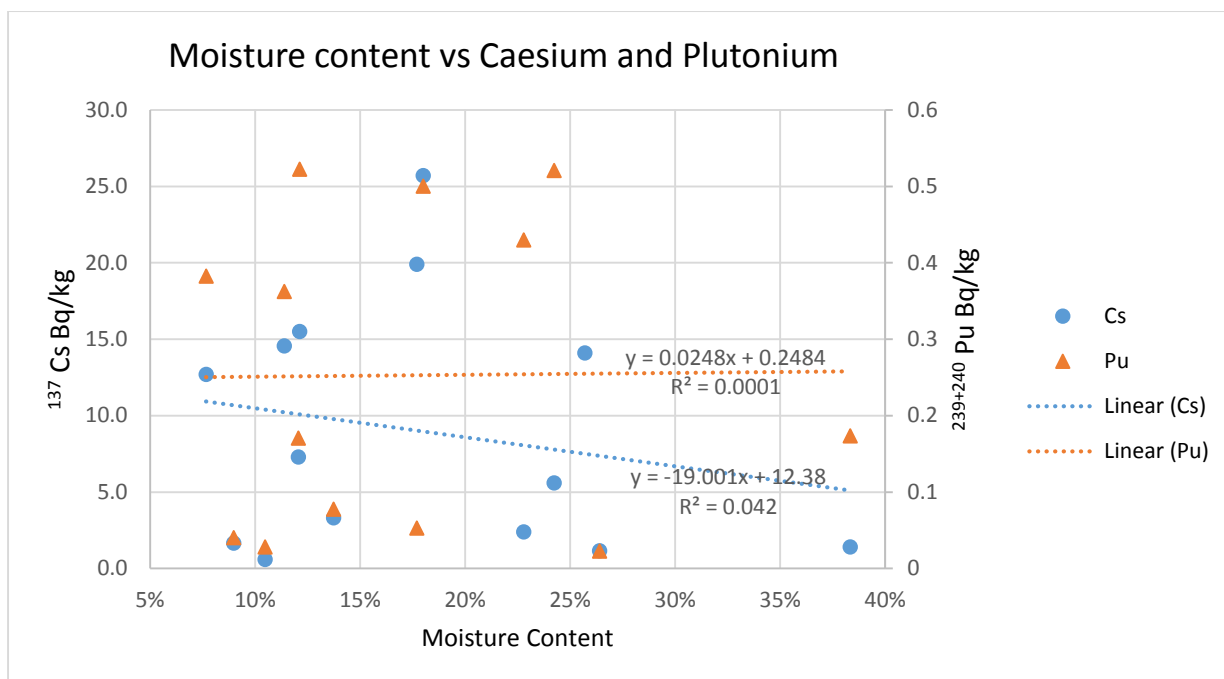


Figure 3.7: Soil moisture content compared with Caesium and Plutonium content.

3.5 Loss on Ignition – Organic Contents

The loss on ignition (LOI) represents the amount of organic matter present in the soil. The soils were found to have between 10% and 20% organic matter, while the dry algae collected from the LFBG creek was not measured due to assumptions that it will be near 100% organic and the sample will have been lost. There was limited variation within this (Figure 3.8). There is no apparent relationship when comparing the most westerly site (Evans Lookout) to the most easterly (Forest Brook). There appears to be a relationship between the organic content and the fallout retained in the soil, although this is statistically weak with a fit of $r^2 = 0.25$ for Plutonium and $r^2 = 0.25$ for Caesium (Figure 3.9).

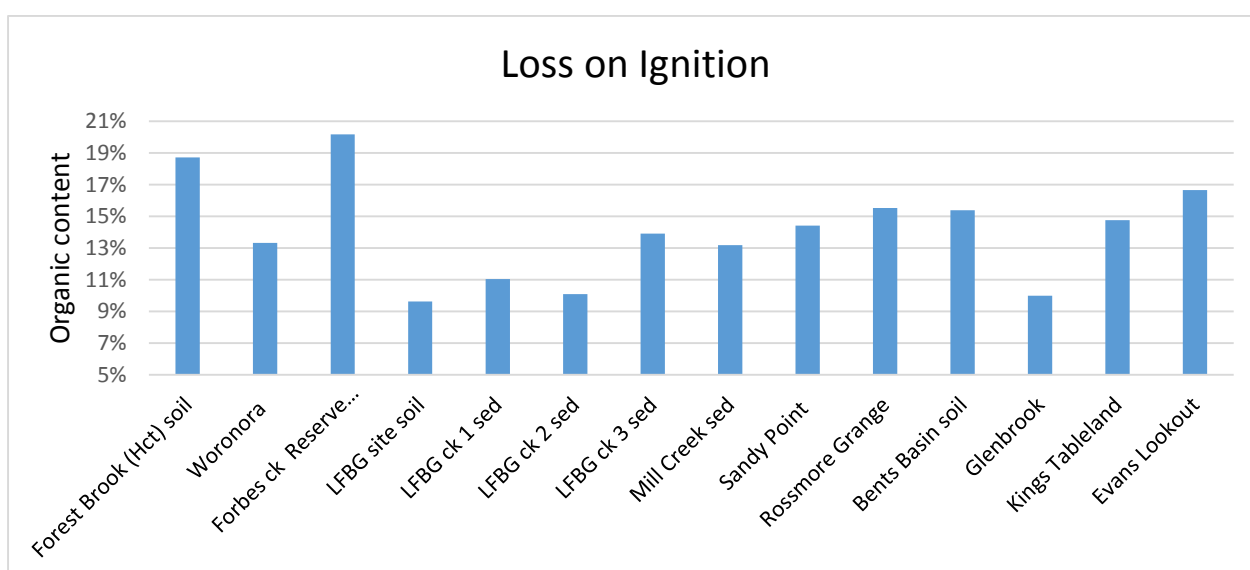


Figure 3.8: Loss on ignition representing organic content of soils.

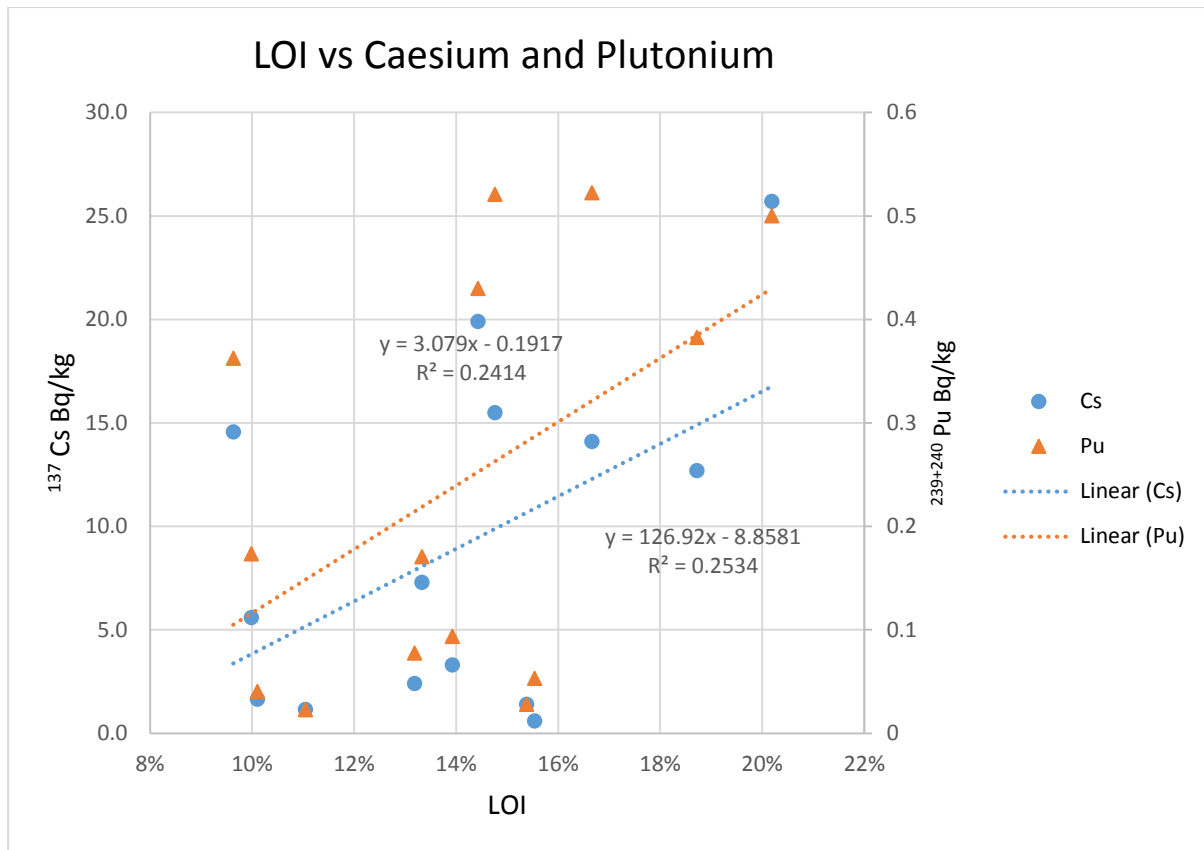


Figure 3.9: Organic content plotted against the Caesium and Plutonium of the soil.

3.6 Grainsize

Grainsize varied significantly between the sites. The initial grainsize measurements separated the soil (and sediment) into two fractions; less than 2mm but greater than 63 microns and less than 63 microns. This produced a large range where the less than 63 micron component ranged from 8-45% of the total sample (by dry weight). When compared to the amount of Caesium and Plutonium present this gives a negative linear relationship but with a weak fit ($r^2 = 0.34$ Pu and $r^2 = 0.36$ Cs)(Figure 3.10).

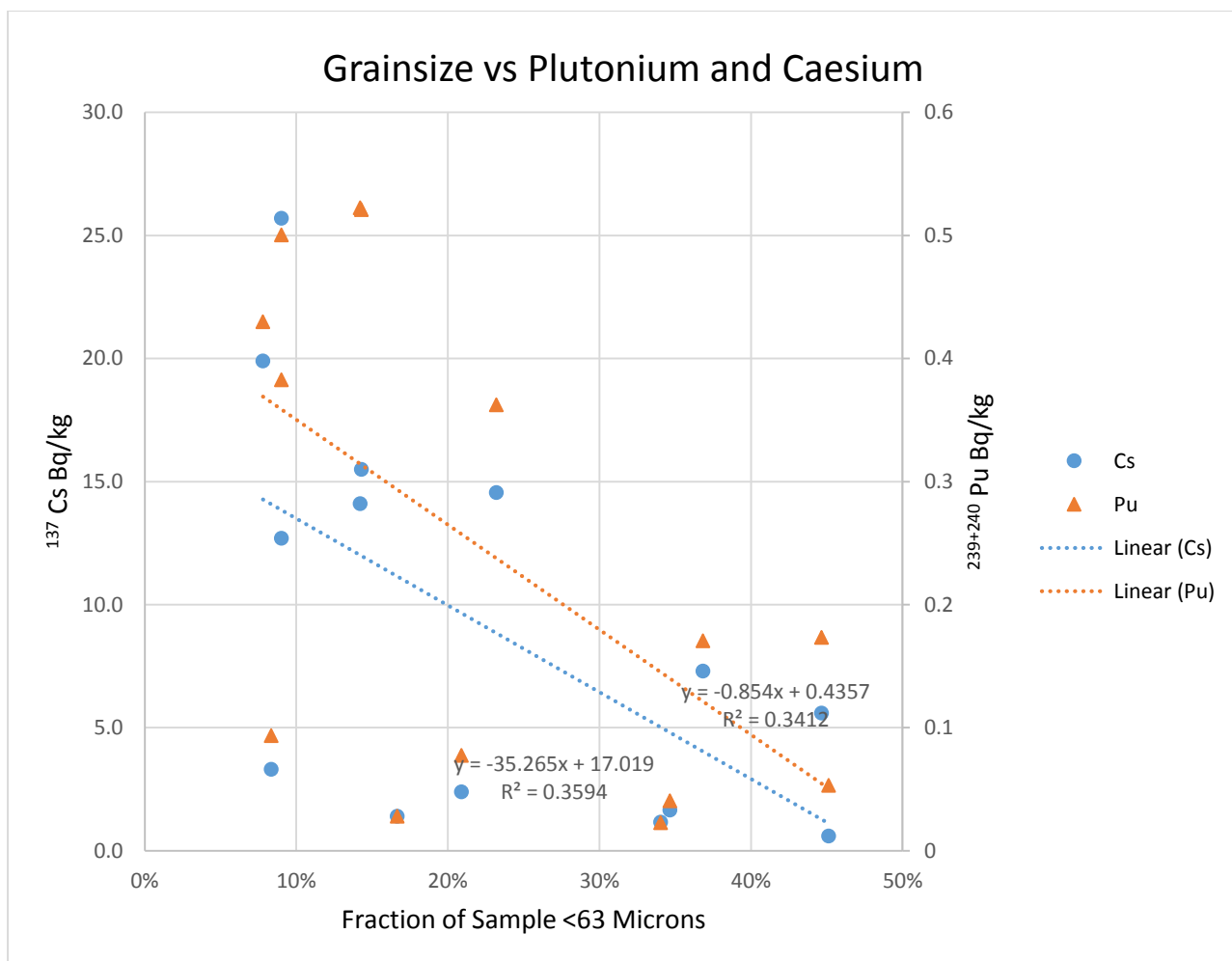


Figure 3.10: Comparison between the size of the <63micron fraction and the amount of radionuclide present.

Figure 3.11 shows the grain size percentiles for analysed samples. The y-axis shows the maximum size of each respective percentile. For example the tenth percentile of the Little Forest Background Soil is 10 microns, meaning that 10% of the sample is less than 10 microns in diameter. One hundred percent of the sample is less than 1000microns (1 millimetre) as they were sieved to 1 millimetre prior to analysis.

The sites show similar numbers for the finest 10% with the exception of LFBG creek 1, which shows much coarser grainsize with a 10th percentile of 53 microns. The finest of the soils was the Glenbrook site with 90% of the grains smaller than 55 microns making almost all of this sample finer than the LFBG creek 1's finest fraction.

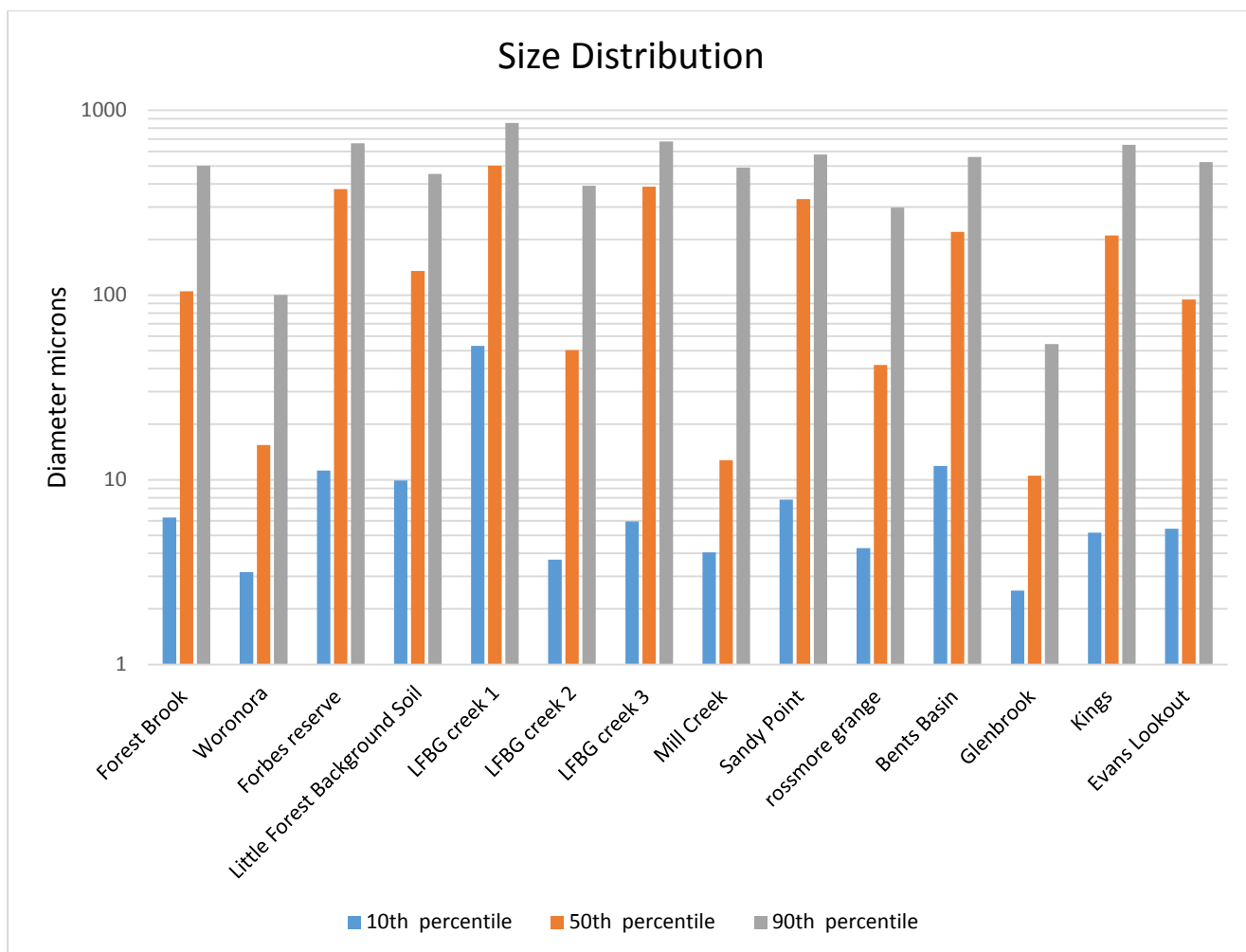


Figure 3.11: Grainsize distribution of the samples.

The method of sieving the samples was not comprehensive as shown in Figure 3.12. The fraction of the samples that was less than 63 microns was greatly underestimated by up to half of what was actually there. Sites such as Glenbrook have shown fine fractions of 88% when using data from the MasterSizer as opposed to 45% when only using the sieve.

The finer grainsizes have shown a higher plutonium activity. When comparing plutonium analysis between the bulk soils and the <63 microns fraction it was found that the percent of plutonium represented within the finer fraction was greater than the portion of the sample that the finer fractions represented. This relationship can be seen in Figure 3.13. Since not all of the fine particles would have been separated from the coarse particles in the sieving process, the calculations resulted in some appearing to have greater than 100% in the <63 micron fraction.

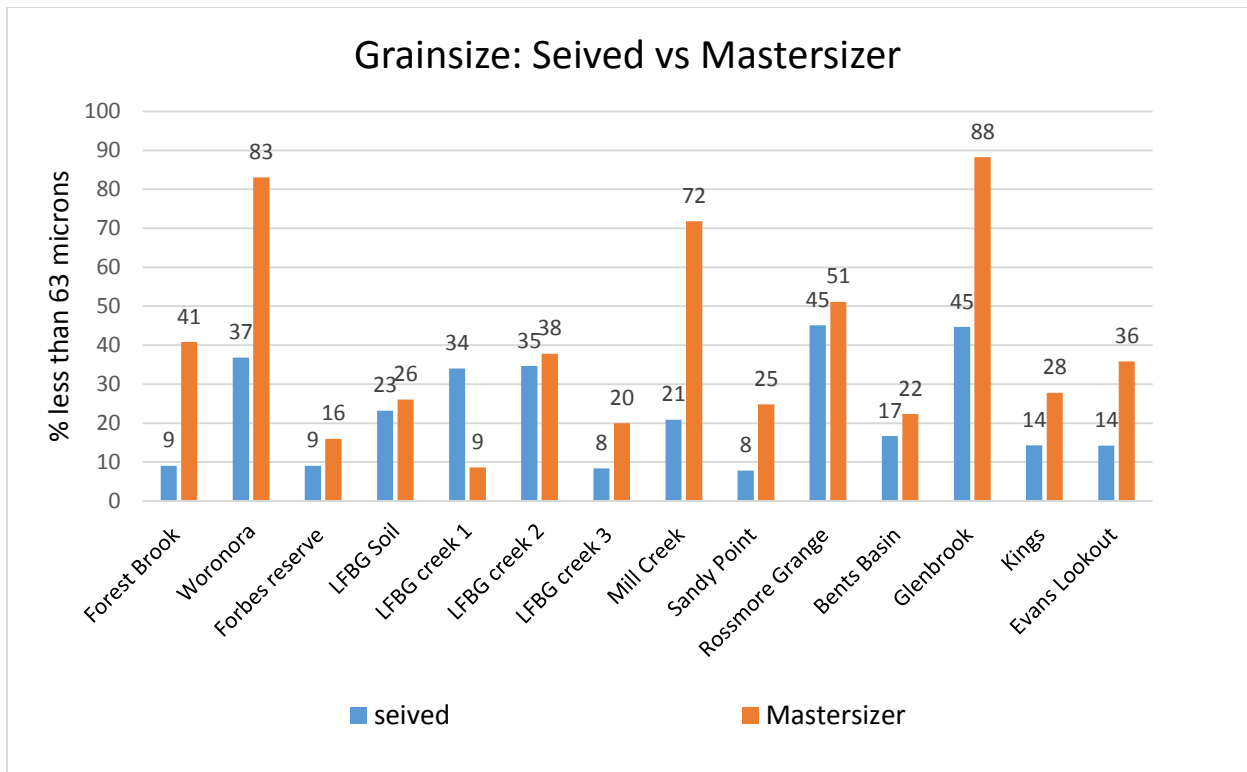


Figure 3.12: Comparison between sieving as MasterSizer grainsize.

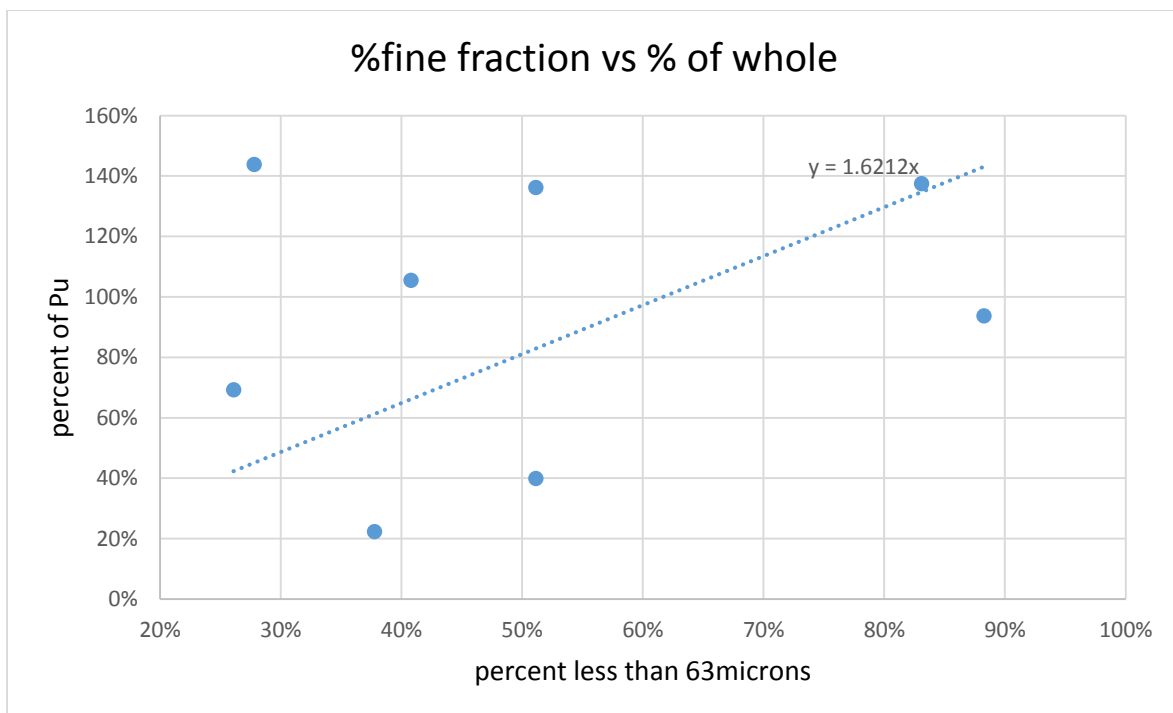


Figure 3.13: Comparison between the percent of the soil represented in the <63 microns fraction and the proportion of the plutonium it represents.

3.7 pH

Each of the soil samples were tested for pH using the standard IOS method. The results showed pH varied from between 4.8 and 6.2 showing that the soils are mildly acidic (Figure 3.14) Woronora shows the lowest pH at 4.8 while Mill Creek is the highest at 6.2. The relationship between the pH and the fallout retained in the soils can be seen in Figure 3.15. This shows that there is no significant relationship between the pH of the soils and the amount of fallout in which they have retained.

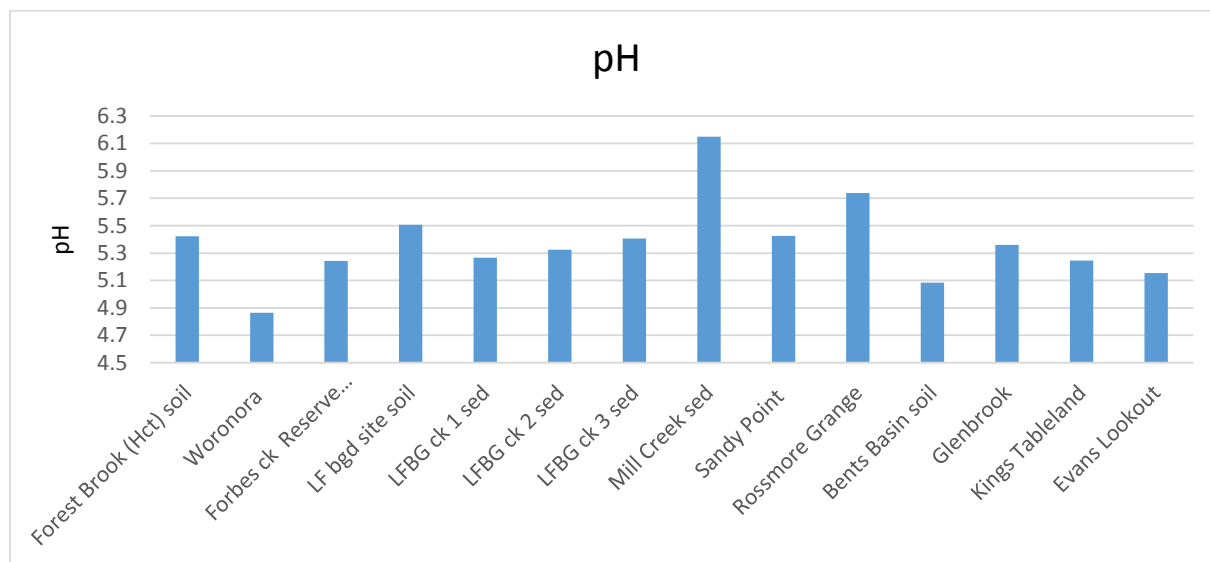


Figure 3.14: pH of soils.

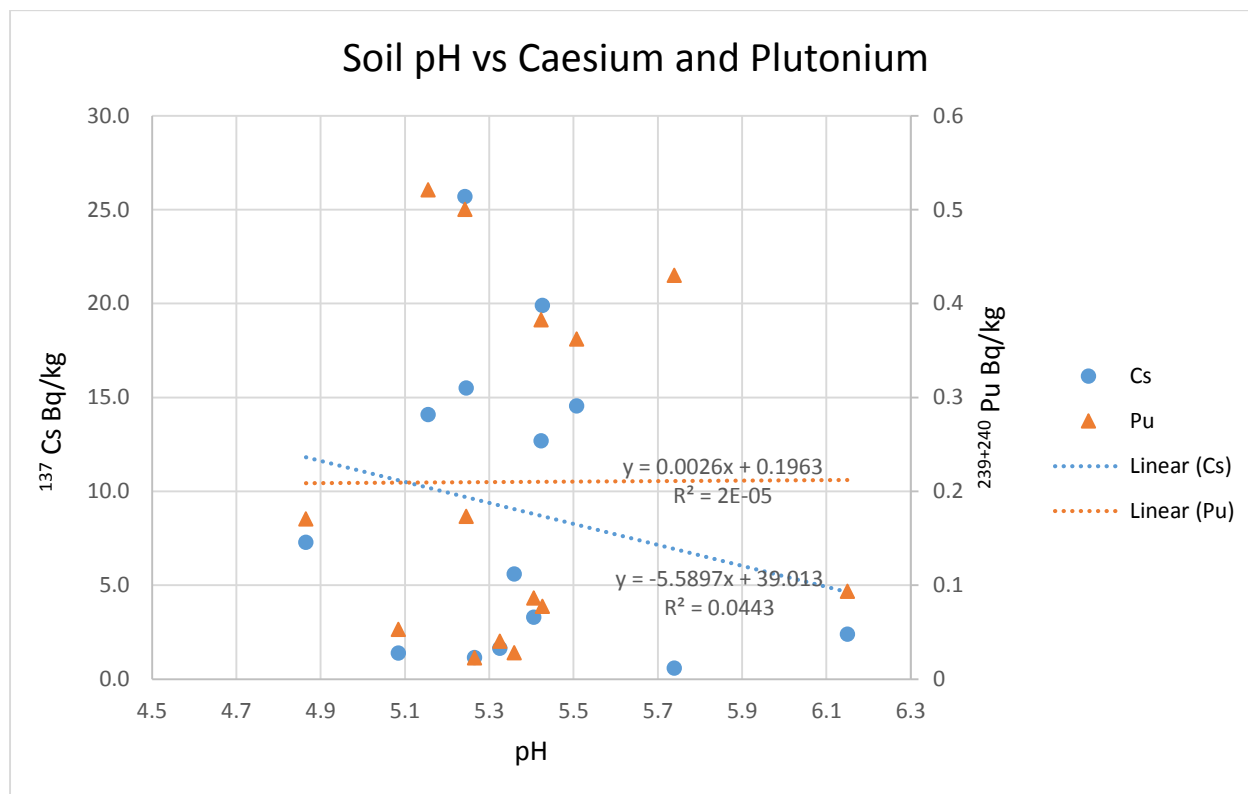


Figure 3.15: Comparison between Soil pH and the Caesium and Plutonium measured.

3.8 Elevation

Elevation was similar for many of the sites with the exception of those located within the Blue Mountains. Elevation was suspected as being related to the amount of fallout received, as higher altitudes will be closer to the plumes in which the fallout was spread. While the results show a general positive trend for plutonium the fit of the trend is weak ($r^2 = 0.37$ for Pu and $r^2 = 0.09$ for Cs) (Figure 3.16).

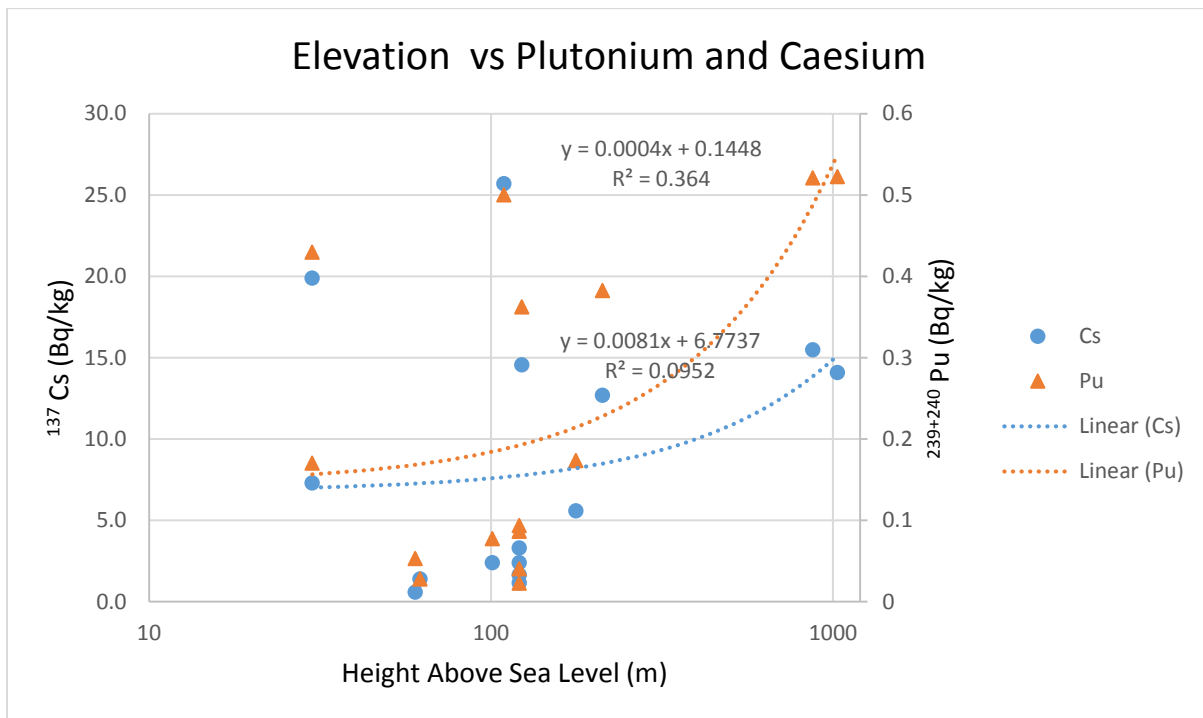


Figure 3.16: Relationship between elevation and fallout. Many sites have similar elevations and this has affected the trends.

3.9 Rainfall

Rainfall data was collected from the closest BOM weather station to the site. This means that they are not specific to the site and that multiple sites will fall within the one rainfall measurement. It is shown in Figure 3.17 that many of the sites have the same measurement for rainfall, this is due to them all being located in the Lucas Heights area. There is no relationship between either of the radionuclides and rainfall with r^2 values of 0.1 and 0.03.

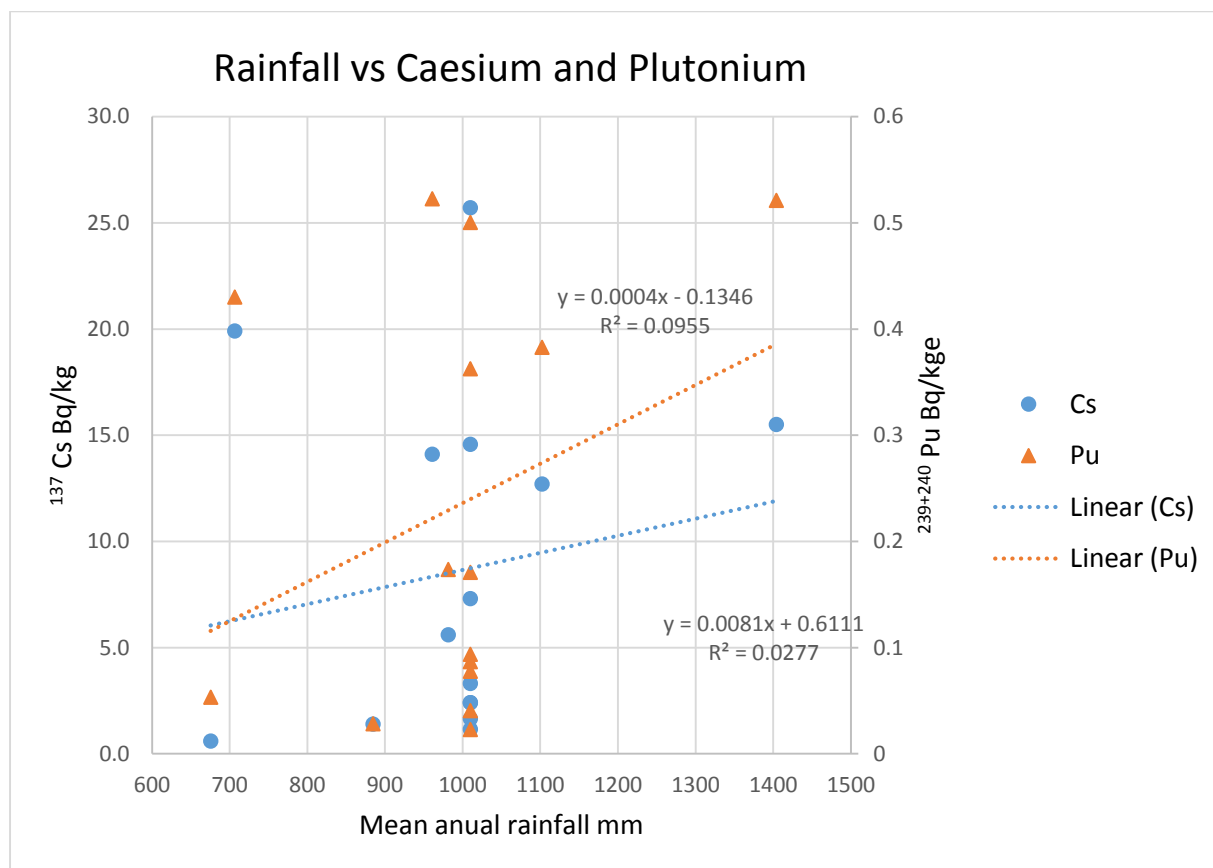


Figure 3.17: Rainfall vs fallout for all sites.

Chapter 4 - Discussion

This chapter discusses the results of the study and the broader context of the importance of measuring radionuclides. The main focus of this thesis was to expand upon existing measurements of plutonium and caesium for the Australian continent by recording measurements for the Sydney area. The use of plutonium as a tracer in erosional studies is relatively recent and consequently measurements are scarce. These measurements will become increasingly important into the future as caesium, which is more frequently used, decays beyond detection limits. The baseline established from this work will assist future studies into erosion, global fallout, the assessment of nuclear facilities and legacy waste sites.

4.1 Plutonium isotope ratios

This study measured the $^{240}/^{239}$ Pu isotope ratio which can be used to assess global fallout sources. The plutonium isotope analysis conducted within this study support the other measurements that have previously been published, and suggest that the $^{240}/^{239}$ Pu ratio of plutonium isotopes for Australia is below the global average (Tims et al., 2013, Child and Hotchkis, 2013, Kelley et al., 1999, Everett et al., 2008). This is shown in Figure 4.1. The lower ranges are measured for test sites at Maralinga and Emu Fields. These are closer to detonation points and as such are receiving short range fallout as opposed to the long range fallout that will have reached the east coast of Australia (Child and Hotchkis, 2013).

The work of Tims et al. (2013) included values from throughout continental Australia. These values were the result of compiling values from several nearby sites to form a representative value (Tims pers com). There is however, little data from NSW, with only one site analysed from the Snowy Mountains included in this study. With no sites for Sydney currently published this project was increasingly important. Following discussion with Dr Tims, it was revealed that an isotope measurement was also taken from the Kings Tableland area but not included within the publication (Tims pers com). As the present study has also measured the Kings Tableland area this result could prove a useful comparison.

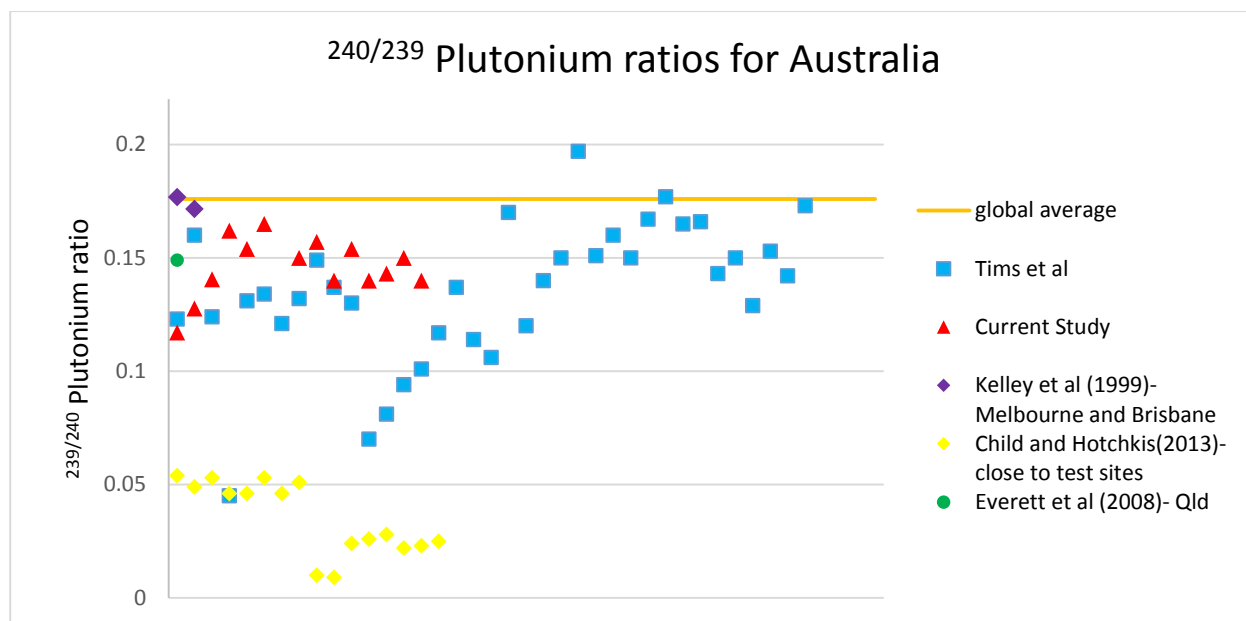


Figure 4.1: Comparison between measured data and published values of $^{240/239}$ Pu isotopic ratios for Australia. The X axis is representative of sample number.

All the results in Figure 4.1 are lower than global average values, which suggests that the atomic weapons tests conducted within Australia have resulted in a unique and regional fallout pattern (Child and Hotchkis, 2013). The consistently lower values reflect the specific sources and isotopic composition of the fallout received by the Australian continent. As such, measurements taken in other parts of the world for erosional studies may not be applicable to the Australian context.

Internationally there has been more work into the measurement of the $^{240/239}$ Pu isotope ratio than there has been within Australia (Green et al., 1991, Kelley et al., 1999). These isotope measurements have been applied to sediments such as those in Lake Ontario (on the USA Canada border) and used to determine whether a processing plant had released plutonium into the lake (Green et al., 1991). The collection of more measurements and establishing a base level for radionuclides dispersed across Australia will enable similar hypotheses to be tested in the future, such as the impacts of any future waste sites.

The first measurement of the $^{240/239}$ Pu isotope ratio was conducted by Krey et al. (1976). This study measured the plutonium ratio of several sites in Australia, recording a ratio of 0.1707 for Sydney (Krey et al., 1976). The current study does not support this value with isotope ratios ranging from 0.11 to 0.16. This discrepancy is likely due to the progress in analytical techniques becoming more accurate. Krey (1976) and Kelley (1999) used a TIMS instrument for the Mass spectrometry rather than an AMS, which is more sensitive. Through the Kelley et al. (1999) study it was shown that the southern hemisphere would have different isotopic ratios to the Northern Hemisphere and the Equatorial zones. Kelley suggested that a mean value for areas between 30-53°S would be 0.185, with a latitude of ~33°S. Sydney would fall into this category. The new data collected in this study does not support this higher than global mean value for Australia, with no values higher than 0.165.

4.2 Sedimentation and erosional studies

It is important in the creation of erosion models that there is a method of testing the accuracy of predictions. Recently (late 1990's onwards) radiometric tracers have been used to assess these outputs, as well as identify the major sources of sediment. Caesium-137 is the most commonly used isotope for these measurements but only has a half-life of 30 years (Everett et al., 2008, Loughran et al., 1990, Simms et al., 2008a). Therefore it is becoming important to find a replacement for ^{137}Cs in erosional studies, particularly considering that over 60% of the caesium inventory has already been lost to decay (Everett et al., 2008). Plutonium has a much longer half-life than caesium-137 and was deposited as a result of the same fallout associated with atomic testing. The correlation between plutonium and caesium is strong (Everett et al., 2008) confirming plutonium as a valid replacement for the future when caesium will begin to fall below detection limits. The current study supports this correlation.

The breakdown of the caesium-137 increases the importance in establishing plutonium values for the sites. This study provides indications as to the levels plutonium present in the Sydney area. This will be important in the design of future studies as the baseline allows estimates of the detection limits required for the analysis. When comparing all the samples from the current study there is a linear correlation between the plutonium and the caesium with a fit of around 87%. This is a strong correlation, though a higher percentage would have been desired. This may be attributed to only analysing 14 samples, the combination of soil and sediment sample, or perhaps a function of the Sydney Basin environment.

For erosional studies it is important that many measurements can be taken in order to establish a pattern. When it comes to the measurement of multiple samples gamma spectrometry requires very little sample preparation but a long counting time. For this study samples were counted for up to 3 days. As the machine can only analyse one sample at a time this becomes either an expensive process with multiple machines, or a very slow one. Though they require more processing, AMS allows many samples to be analysed quickly (Hoo et al., 2011) allowing for larger batch sizes. The downside to AMS is that it is currently much more expensive than gamma spectrometry.

Internationally, plutonium is of greater importance to erosional studies (Alewell et al., 2014, Schimmack et al., 2001). This is in part due to the Chernobyl disaster, which released large quantities of caesium into the European environment during April and May of 1986. At this time of the year much of the Alps were still covered in snow. This deposition of caesium onto snow led to an uneven distribution as the snow melted and the caesium was transported elsewhere. In comparison to this quick and disturbed fallout of the caesium, the plutonium produced from atomic testing was evenly spread and deposited over a greater period of time. This allows plutonium to be a much more consistent tracer for erosional studies (Alewell et al., 2014).

The measurements taken in this study combine with many others to assist our understanding of how the atomic testing of the 1950's and 1960's impacted the world. Caesium-137 does not occur naturally and so all the caesium-137 measured is a result of these nuclear events. The presence of caesium-137 has become a distinct chronological marker allowing for the

dating of sediments as pre or post atomic era. As the caesium-137 was only deposited over a period of 14 years it is able to represent a very small window of history. This 14 year period is the time that it took for the caesium-137 to be released and deposited into the environment (Rember et al., 1993).

4.3 Implications of the baseline data

This study has provided a collection of baseline data for the Sydney Region. This region includes Australia's only nuclear reactor, the OPAL reactor which is used for both research and the production of radio-pharmaceuticals. This reactor replaced the HIFAR research reactor in 2007. The region also includes the Little Forest legacy waste site (formally known as Little Forrest Burial Ground). At this location the former Australian Atomic Energy Commission (predecessor to ANSTO) buried low-level radioactive waste in multiple trenches excavated into the clay surface soils above a shale lens in the bedrock (Payne et al., 2013). Due to the presence of a potential source of radionuclide contaminants close to the major population centre of Sydney it is important to establish baseline levels of radionuclides. These studies could also be useful if a significant global event (such as the Fukushima incident) released radioactivity into the environment.

Implications for the Little Forest Legacy Site

The Little Forest Legacy Site (LFLS) is located in the vicinity of the ANSTO site at Lucas Heights, Sydney. It was used between 1960 and 1968 for the disposal of radioactive waste from the Australian Atomic Energy Commission. This waste includes several grams (exact value unknown) of plutonium along with other radioactive sources. When compared to other legacy waste sites (for example those in the USA where up to 200kg of plutonium was disposed) this site is significantly less hazardous (Payne et al., 2013).

Recently there have been extensive studies on the site to measure radionuclide distributions at the Little Forest site (Harrison et al., 2011, Hughes et al., 2011, Payne et al., 2013, Twining et al., 2011). These studies collected surface soil measurements for plutonium concentrations. Several surface soil samples from the immediate vicinity of the trenches containing the disposed plutonium indicated the presence of $^{239+240}\text{Pu}$ above 100 Bq/kg. The elevated plutonium in the soils can be attributed to a "bath tubbing" effect where by the trenches fill with water during periods of high rainfall allowing the plutonium to flow over the top (Payne et al., 2013).

However, other soil samples from around the perimeter of the site demonstrated levels of $^{239+240}\text{Pu}$ of less than 0.4Bq/kg (Payne et al., 2013). Part of the justification of the present project was to establish whether these lower levels could be attributed to contamination derived from the trenches or could be representative of global fallout. The current study has found that the range of $^{239+240}\text{Pu}$ values for the Sydney Region is 0.02 to 0.52 Bq/kg with a mean of 0.24 (standard deviation 0.2 for <63 microns fraction). This means that the lower samples from the Little Forest Site are within the range of plutonium concentrations occurring at sites with no radioactive waste disposal. However, the AMS measurements in the current study only determined the plutonium values as concentrations of the finer fractions, whereas the LFLS were for bulk soil samples. The bulk soil samples analysed for

this study show concentrations of 0.04 to 0.20 Bq/kg, which is below the levels that have been measured at Little Forest, although it may indicate that a substantial proportion of the plutonium in the more distant samples results from fallout rather than originating from the trenches.

Alluvial sediment samples have been measured for a separate study from an ephemeral creek draining the LFLS. This site has plutonium values in the range of 0.6-1 Bq/kg, which is much higher than the bulk value for the site measured for this study. The creek that does not drain the site had an activity of 0.076 Bq/kg (bulk), an order of magnitude smaller than the creek which receives the LFLS site drainage. More studies are required and are being undertaken to determine if these elevated results are caused by the legacy site.

Isotopic ratios have previously been analysed to determine the source of the plutonium in the LFLS soils. These values are lower than those found in this current study (Figure 4.2) (ANSTO in preparation). Lower values suggest that there is more plutonium-239 present than would be expected for regional Sydney fallout. These values are also lower than average when compared to the study conducted by Tims et al. (2013) indicating they are different to most of Australia but are still within the lower limits of the range for the continent.

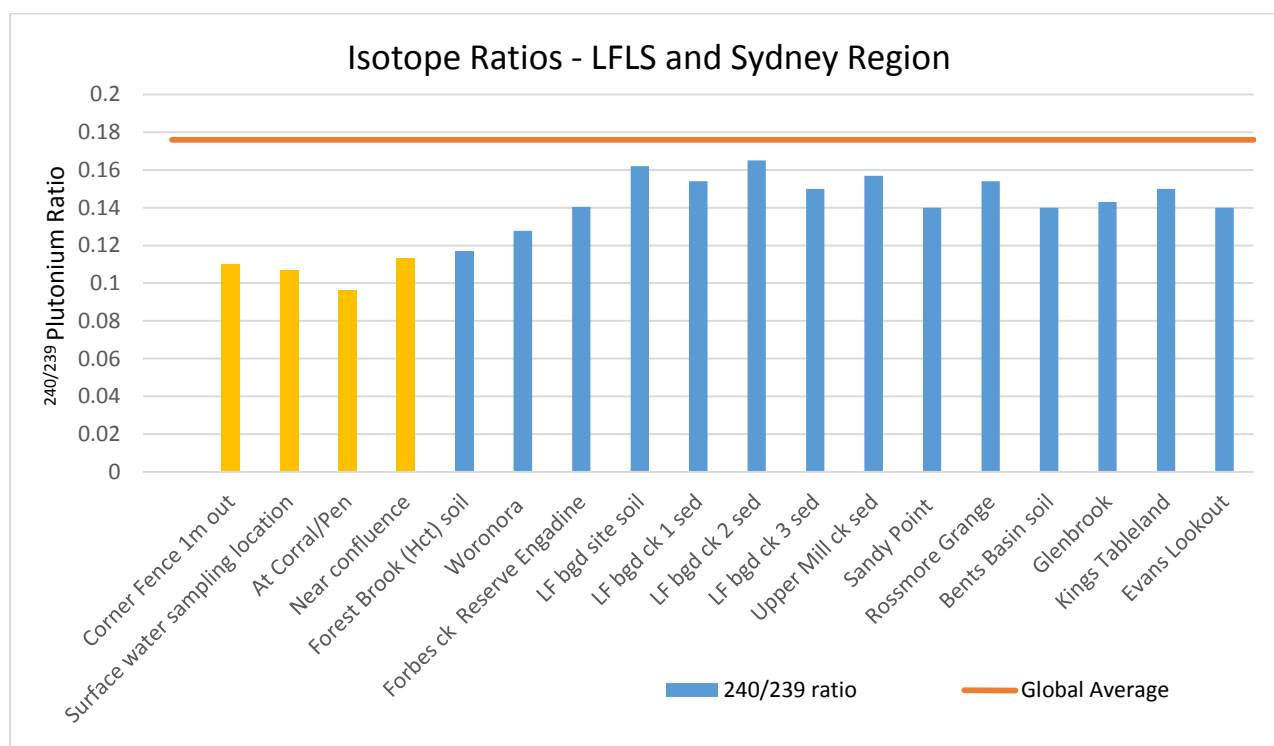


Figure 4.2 Comparison between ²⁴⁰/₂₃₉Pu ratios near the Little Forest Legacy Site (yellow) to the current study (blue) and Global average (red line).

4.4 Variability in Pu and Cs activity in the Sydney Basin

During analysis further information about some sites was discovered indicating that they may not have been as ideal as once thought. Rather than abandon these samples, they were analysed along with the others. These samples have shown significantly lower levels of ^{137}Cs and $^{240+239}\text{Pu}$ than the others. Potential reasons for this are discussed below.

Rossmore Grange

The Rossmore Grange site features extremely low caesium and plutonium activity. This may be due to disturbances within the site, which would have resulted in dispersion of radionuclides deposited by fallout. The Rossmore Grange site was originally allocated as part of a 700 acre land grant to William Emmett around 1818 and was farmed up until at least 1890. The family is still reported as owning the land until at least 1940, after which it appears to have been abandoned. In 1978 it was purchased by the Liverpool City Council and used as a public park and protection for areas of the now threatened Cumberland Plain vegetation (Smith et al., 2006). During the time gap between the atomic testing and depositing of fallout in the 1950's and 1960's and the point in which it became Council land many disturbances might have occurred from both natural and anthropocentric circumstances. These are unmeasurable, and while the best care was taken to sample from the least disturbed section of the site, the level of disturbance remains unknown. Thus the low levels of ^{137}Cs and $^{240+239}\text{Pu}$ activity at this site are believed to result from significant soil disturbance.

Little Forest sites

Caesium and plutonium activities for the small creeks used as the Little Forest Background samples were much lower than from other sites. The disturbance to the Little Forest sites is much more quantifiable than that of the Rossmore Grange site, although it was only discovered after analysis of the samples had already begun. Aerial photography (Figure 4.3 and Appendix 2) of the region shows that at some point between the 1970 and 1984 (Figure 4.3) extensive clearing was undertaken within the area. This disturbance is not directly placed over the soil sample site (which was closer to a cliff and would have likely been too dangerous to clear). It is however likely to have had a large impact on the sediments in the creeks as these drain directly from the disturbed parts of the site. One possible explanation of the Pu and Cs values that have been observed within the sediments measured at this site is that as erosion increases more sediment moves down the creeks. This makes it less likely that the sediment is from the upper layers of the soil that have collected the fallout.



Figure 4.3 Aerial Photography of the Little Forest Back Ground Site (red) with cleared patches nearby.

4.5 Plutonium Variability

Collecting from varied locations within a site aimed to reduce the impact of heterogeneous fallout patterns caused by features such as canopy cover and ground surface differences (Hoo et al., 2011). As spatial variability is not easily accounted for, a range of plutonium and caesium values were recorded.

A mean value of 0.24 Bq/kg was measured for the plutonium in the Sydney Basin, though the plutonium reported in the results section of this report represents only the plutonium in the sieved fraction. If this is to be adjusted to the whole there would in fact be a much lower concentration of plutonium. If it was assumed that 100% of the plutonium was contained within the less than 63 micron fraction (a poor assumption) then the average plutonium would result in a concentration of 0.07 Bq/kg. This value is on the lower end of the concentrations reported by Tims et al. (2013), and as the plutonium is not totally contained within this fraction this number is an underestimate by up to 50% as shown by the alpha analysis of the bulk soils. Even if the 0.07 Bq/kg deviates by this much it will still create a value of 0.14 Bq/kg. The values for plutonium (239 + 240) previously measured for the ACT average to a value of 0.356 Bq/kg (range from 0.055 to 1.003) (Tims et al., 2013). This makes Sydney appear to be lower than the average for sites measured in the ACT. Other sites measured around the country present lower plutonium concentrations. Sydney's activities appear higher than the measured plutonium in the Northern Territory and Western Australia. Sydney appears to have a similar concentration of plutonium to Queensland (Tims et al., 2013). This may be due to weather effecting the fallout deposition. The ACT predominately is influenced by weather systems from the west heading east, as is Sydney. However, Sydney is also influenced by east coast lows, which will bring air masses from the south-east, away from the British test sites.

Plutonium attaches to organic matter in the near surface soils, and in the event of a fire this material may be redistributed. It has also been reported that forest fires may resuspend radionuclides (Hoo et al., 2011) causing their removal or movement by other factors. Bushfire occurrence has not been a factor taken into account for this study despite the areas being areas of high fire risk. This may explain the inconsistency of the plutonium concentrations between some of the areas.

4.6 Caesium Variability

The caesium results vary similarly with the plutonium results for the sites studied. The alluvial sediment samples all contained a low level of caesium while some soils also had low levels. The Forbes Creek Reserve has a high level of caesium and as such was rerun four times to check for analytical errors. Despite this, the level detected remained at 25 Bq/kg. There appears to be no environmental factor from the ones measured that influences this. Currently the best explanation is that the housing developments surrounding the reserve have somehow impacted the erosion/deposition of the caesium, causing it to have an elevated concentration for the site that was sampled. This site was different from others as it was not the top flat section of a hill, but a flat section approximately mid-way down a steep slope. There can be considerable variability in ^{137}Cs fallout due to rain shadowing and small-scale run-on-runoff processes at the time of deposition (Wallbrink and Murray, 1996). As

deposition was more than 50 years ago these factors have not been able to be measured. The value of 25 Bq/kg is higher than other reported values, but not excessively, Simms et al. (2008) reported a caesium concentration of 17 Bq/kg for a site to the south of Sydney (Simms et al., 2008a).

The Rossmore site demonstrates extremely low caesium levels, suggesting that it has been disturbed since the atomic era. This has already been explained earlier in the site evaluations section of this discussion.

The variations in the caesium values found in this study are supported by other caesium values that have been collected in the region. Values of between 1 and 17 Bq/kg have been recorded from erosional studies in the area (Simms et al., 2008b). The acidic nature of the soils creates an environment where the caesium is least mobile (Loughran et al., 1990). This gives us confidence that the caesium measured represents the original fallout and that little has been removed through chemical actions.

4.7 Caesium and Plutonium activities, as related to environmental factors

Grainsize

The general hypothesis tested for grainsize is whether finer soils will have higher concentrations of caesium and plutonium. The relationship between the grainsize and the fallout initially appeared to demonstrate that the radionuclides are not directly concentrated within the finer particles of the soil. This however does not appear true with the proportion of the total plutonium in the fine fraction, being greater than that fraction as shown in the alpha analysis of the plutonium. If however we are to assume that the entire Sydney area would have initially received the same amount of fallout, and that this fallout was concentrated within the finer soil particles a dilution effect would have occurred within the soils that had a high percentage of fine particles. This is a slightly misleading result of expressing the values as Bq/kg for the finer fractions.

Example: fallout = X

X in 10kg = X/10 Bq/kg

X in 100 kg = X/100 Bq/kg

Therefore the activity would appear smaller, but this could be a result of more fine particles.

It was not anticipated that the sieving process would be able to separate 100% of the less than 63 micron fraction from the soil. However, the huge discrepancy between the sieved data and the MasterSizer data was surprising. When the data is adjusted from the <63 micron fraction using the MasterSizer there is a much greater agreement between the alpha spectrometry values and those obtained from the AMS. This supports that the plutonium is indeed concentrated within the fine particles of the soil.

Moisture

Moisture content does not appear to have any significant impact on the retention and presence of the radionuclides measured, as no correlation between moisture content occurred. The moisture content of soils is highly variable and dependent upon the recent rainfall, antecedent moisture, grain size, aspect and exposure, geomorphic position and vegetation. These factors vary within both spatial and temporal contexts. This is especially true for the samples that were taken from the upper levels of the soil and would have been relatively free from ground water impacts. The rain will have influenced the moisture of the soil as on the previous night, and during some of the collections it had rained.

Loss on Ignition (LOI)

Caesium was expected to have a negative relationship with the organic content in the soils due to increased mobility in organic soils. Therefore it is surprising that there was no observable correlation between the organic content of the sample and the amount of radionuclides it contained. This may be due to the small range of organic contents that were recorded for the samples, between 10-20%. It may be that this range was not large enough to detect any patterns in the relationship between organic matter in the soil and the retention of radionuclides. Furthermore any organic effect, if it occurred, would need to be significant enough to overcome the other controlling factors (such as elevation, rainfall, erosion history, etc.) and this is unlikely given the small size and limited diversity of the present sample set.

pH

Not surprisingly the pH varied very little between the sites with all sites shown to be acidic in nature. While the pH is reported to influence the retention of fallout in soils, favouring acidic soils, no trend was detected. This may be that because the range of pH was very small and the variability in the radionuclide concentrations very high that no pattern was evident.

Elevation

Due to the majority of the samples having a similar elevation it is unclear if a pattern between the elevation and fallout exists. More samples particularly those with elevations between 200-1000m are required to increase the insight.

Rainfall

Site specific rainfall data was not available. This is due to the close proximity of many of the sites as well as the low density of the weather stations. As many of the sites were close to Lucas Heights they all have a rainfall from the same weather station measuring 1010mm mean annual rainfall.

The overall measurement of the environmental factors does not show any correlation to the amount of caesium-137 and plutonium-240+239 present. This is likely due to the large range of other factors that will be influencing the retention and mobility of these elements.

Soil types

The amount of plutonium-240+239 and caesium-137 in the samples may have been influenced by the soil types. The highest activities were found to correspond with the siliceous and earthy sands (Evans Lookout, Kings Tableland, Forbes Creek Reserve and Forest Brook). However, the lowest plutonium concentration was also found on this soil type (Bents Basin). The Yellow Podzolic soils (Rossmore Grange, Sandy Point, LFBG) generally have lower levels than the Siliceous Sands, though they are also higher than the Bents Basin and Glenbrook samples. This may indicate that the different soil types will have different fallout retentions. The formation of soil may also differ between these sites. If soil is forming faster than the fallout was deposited on the surface of the soil in the 1950's and 1960's it may have been buried further into the profile than the 5cm sample could detect.

4.8 The analytical procedures

The main issues that affected the analysis of the samples were caused by the chemistry of the samples and the complexity of the chemical separation process. Soil is a complex sample matrix that includes many different parameters which often interfered with the separations and analysis. The AMS processing proved difficult as there was a large amount of iron present in all samples as was shown by large volumes of iron hydroxide forming during plutonium separation. This made it difficult to remove the residual Cl^- from the large volume of iron hydroxide co-precipitate which had formed (limits as to the size of the centrifuge tubes made washing with equal amounts of precipitate to water difficult). This problem would have been greatly reduced if 1g of ashed sample was analysed as opposed to the 5g that were used. Based on the results and analytical efficiencies, the amount of Pu measured by the AMS was well within detection ranges and therefore it would have been practical to use a smaller sample mass (e.g. 1 g).

The Sydney Basin has been significantly cleared since the arrival of European settlers. This made the selections of sites difficult. Initially the sites were targeted based upon geology, with ideal sites being situated above areas of shale. Shale was targeted as the soil above it would have been much finer and therefore more susceptible to radionuclide collection and retention. These soils are also much more fertile than the sandstone sites leading to almost total clearing for the use of agriculture and housing. As a result only one site, Rossmore Grange, was found to be acceptable in its limited clearance (though found later to be disturbed) and above the shale. All the shale on the eastern edges of the basin has long been cleared and is now home to the housing estates of the Sutherland Shire. To the west where the majority of the bedrock is shale, parks and nature reserves are scarce as the land has been cleared to be used for agriculture, grazing and more recently housing. As a result of this lack of appropriate shale based soil, sites were chosen based upon the amount of disturbance they had received. It became clear that there is no such thing as the perfect site with the undisturbed sites of the eastern coast having little or no soil.

There is also the possibility of some modifications to the sampling program to take into account that the radionuclides may penetrate deeper into the soil. When the sampling procedure was proposed it was determined that the top five centimetres of the soil profile will have contained the majority of the fallout radionuclides. Other studies consulted after

analysis indicate that while the top 5cm can contain up to 77% of the plutonium in the soil that it may also be as low as 25% and that the radionuclide fallout can penetrate much further into the soil (Hoo et al., 2011). This may mean that the total $^{239+240}\text{Pu}$ and ^{137}Cs inventories were not measured.

The process of sieving the samples to the 63micron fraction proved a benefit to the study as a whole. While the alpha spectrometry has shown that some of the plutonium was not present in the fine fractions, it did show that there was more plutonium attached to these finer particles than the larger ones. Though this effect was not investigated for caesium in this study(due to the large amount of time to measure samples), it can be assumed that since caesium and plutonium behave similarly (Hoo et al., 2011) that there is likely to have been the same concentrating relationship for the Caesium. This became important in the analysis as some of the samples came close to the detection limits for the ^{137}Cs . It is probable that without the sieving process, concentrations would not have been quantifiable.

4.9 Summary

This study supports others in indicating that the fallout received by the Australian continent is not in line with global fallout and that $^{240}\text{Pu} / ^{239}\text{Pu}$ isotope ratios in the Sydney Basin are around the 0.15 range rather than the global mean of 0.176 or the southern mean of 0.185. The local distribution of these isotopes is quite variable, which is in part due to the amount of disturbance that has occurred in the Sydney Basin, with a recent history of land-clearing, agriculture and urbanisation.

Plutonium values for the Sydney region appear to be much lower than others measured for the ACT and other parts of Australia, though this may be a function of study design. Caesium values are highly varied yet are consistently higher than other results for caesium in nearby catchments. These findings may be related to the behaviour of the plume due to the wind patterns at the time of weapons tests, the types of soils or the history of land-usage in the sampled sites.

Chapter 5- Conclusion

The primary aim of this study was to establish a baseline of plutonium and caesium in soil and sediment samples from the Sydney region, adding to the growing number of such measurements for the Australian mainland. This was to be achieved through three types of soil analysis

- Caesium concentrations assessed through the use of gamma spectrometry
- Plutonium concentrations assessed through the use of Acceleration Mass Spectrometry(AMS) and alpha spectrometry
- Plutonium ($^{240/239}$ Pu) isotope ratios also measured through the use of AMS

The second aim of this study was to establish a baseline level for fallout Plutonium and Caesium, enabling the Little Forest Legacy Site to be compared to its surrounding areas. This baseline has shown that the values for plutonium in soils and sediments range from 0.02 to 0.52 Bq/kg, while the caesium ranges from 1.2 to 25.7 Bq/kg, with the average for soil being 11.7 Bq/kg and for sediments 2.1 Bq/kg. This indicates that the Little Forest site does have higher plutonium levels than the surrounding areas, though these are a result of a combination of the waste and the fallout. The range of these values is likely attributable to fluctuating levels of deposition due to atmospheric factors such as wind and cloud cover as well as physical factors such as geomorphology and vegetation.

The third aim was to establish a $^{240/239}$ Pu isotope ratio for the Sydney region, as there are no reliable published values currently available for this area. The study confirmed the works of other authors who have suggested that Australia has a significantly lower ratio than the global average. This study found that a value of 0.15 would more suit the Sydney area, rather than the accepted global mean of 0.176.

Caesium has become commonly used for the analysis of erosion, where soils with lower than average caesium have experienced erosion while those with higher have experienced deposition or sedimentation. The local reference point becomes extremely important in these studies. The establishment of a baseline will allow for the evaluation of the quality of the reference point. Though the values measured in this study are variable they show the range in which scientists conducting erosional studies could expect.

The environmental factors of pH, organic content, moisture, rainfall and elevation measured appear to have no effect upon the concentration of caesium or plutonium measured in this study. This is likely due to the complexity and variability of soils. There appears to be a correlation between the grainsize of the soils and the concentration of the radionuclides, with the finer soils having higher activity.

5.1 Further study and recommendations

This study was limited by time. While the methods are often straightforward they take a long time in the laboratory, especially the preparation for AMS. This study could be greatly expanded in order to create a more comprehensive baseline simply by the collection and analysis of more sample sites. Collection of data in a grid like pattern as opposed to a line would allow the use of GIS (Geographic Information Systems) to create a model of the area predicting other sites. With the limited number (14) samples that this study included, extrapolation into a computer model would not have been statistically viable.

- Further studies will be able to apply the values recorded to both erosional and environmental contamination projects.
- Soil depth profiles can be analysed as to confirm if the majority of the plutonium was in fact contained within the top 5cm

Analytically the only recommendation that can be offered is that 5 grams of ashed soil was far too much for the AMS processing. The inclusions of this much soil caused many difficulties in the processing often adding several extra days to analysis times. The reduction of this to 1 gram would have eliminated some of these problems.

Further analysis into the uranium component of these samples is currently underway, but is not included in this thesis due to time restraints. The uranium will be used for similar purposes as the plutonium was for the present study.

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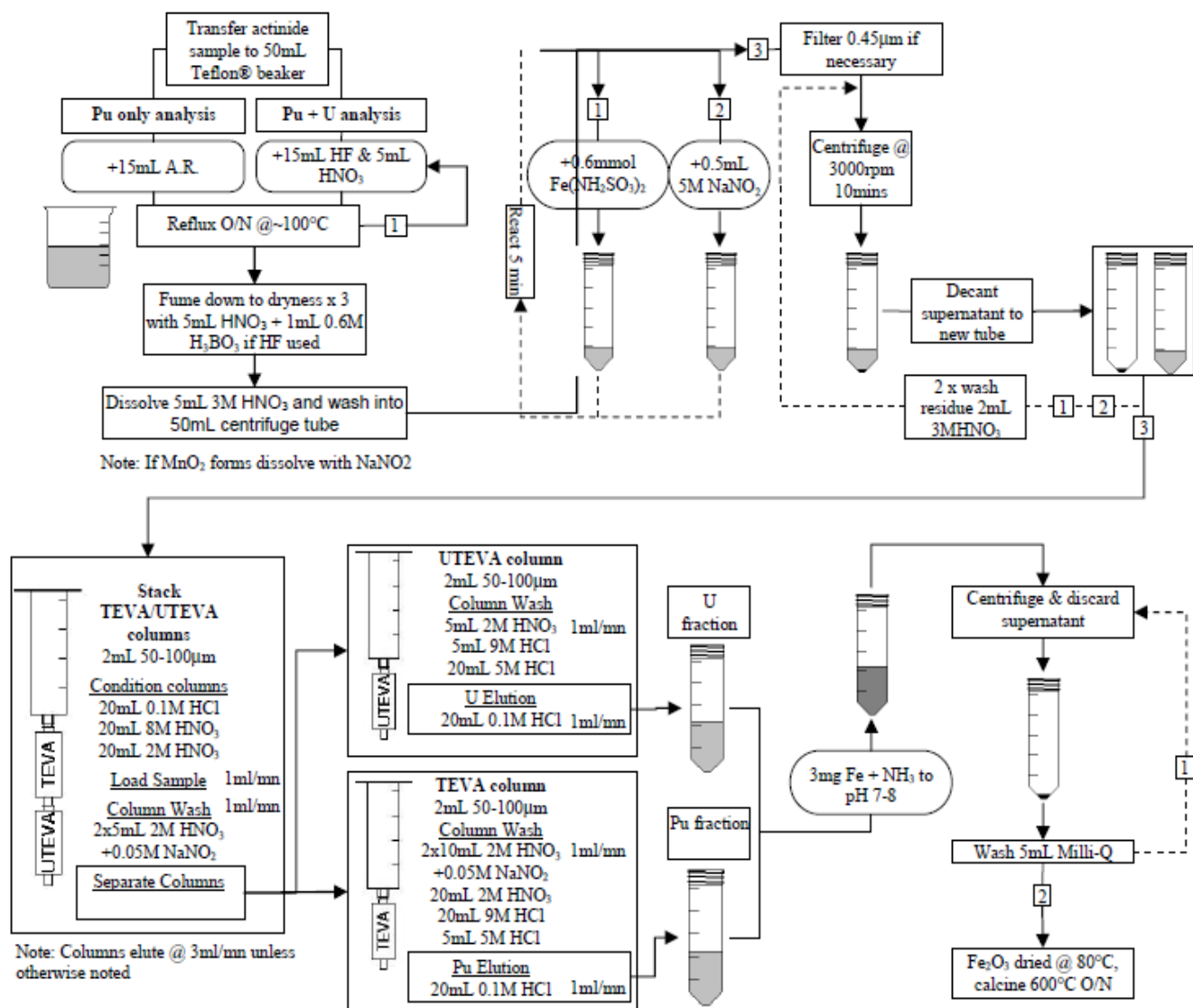
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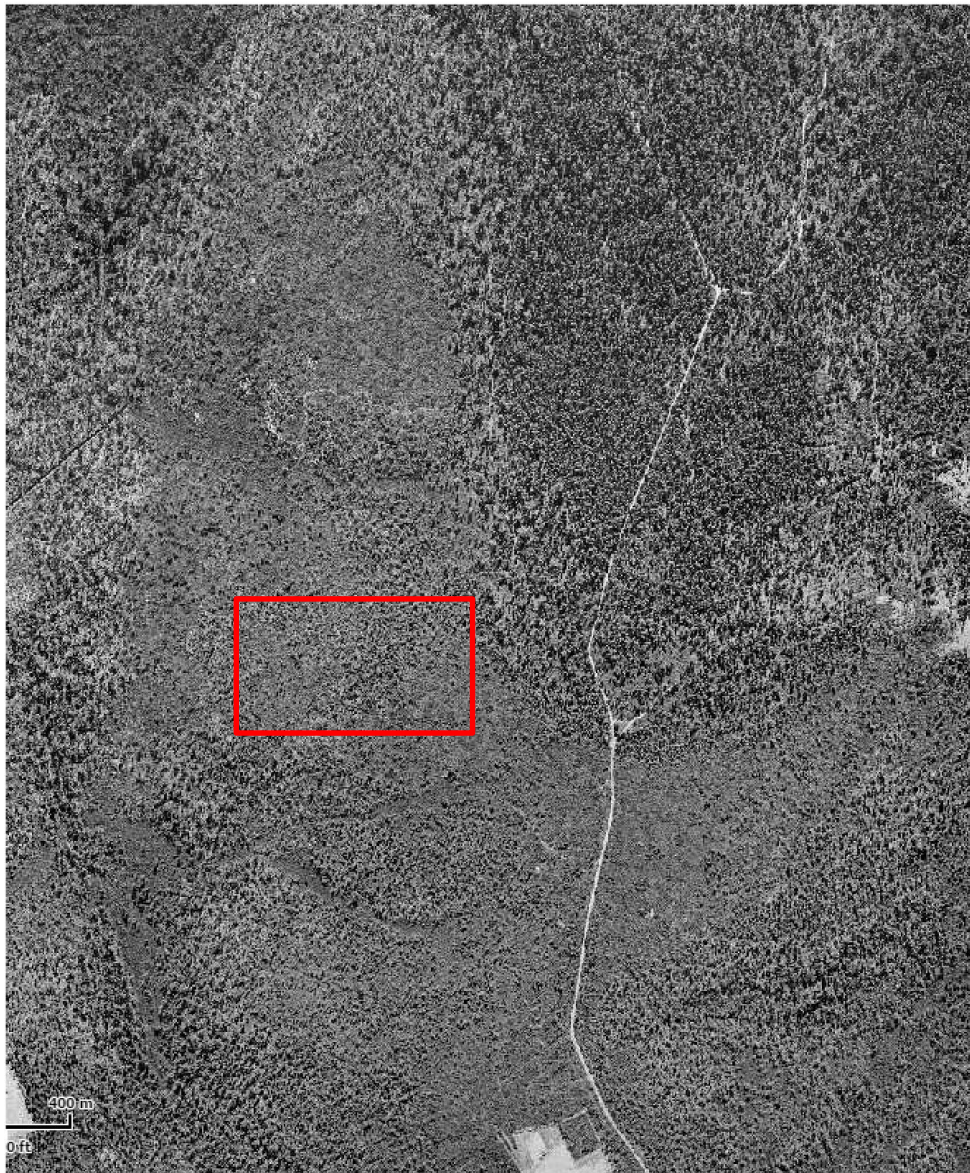
Appendix 1

Purification of actinides flow chart- Plutonium and Uranium from soil.



Appendix 2

Aerial photographs of the Little Forest Legacy Site. Approximate site location is marked in red boxes. Combined map of the photographs is available from the Southerland Shire Library website http://www.sutherlandshire.nsw.gov.au/General/Shire_maps .



1955 Aerial Photo © LPMA

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