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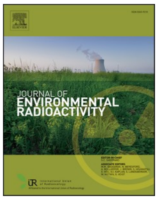

Radionuclide distributions and migration pathways at a legacy trench disposal site

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ABSTRACT

This paper examines the distributions of several anthropogenic radionuclides ($^{239+240}$Pu, ^{241}Am, ^{137}Cs, ^{90}Sr, ^{60}Co and ^{3}H) at a legacy trench disposal site in eastern Australia. We compare the results to previously published data for Pu and tritium at the site. Plutonium has previously been shown to reach the surface by a bath-tubbing mechanism, following filling of the former trenches with water during intense rainfall events. This has led to some movement of Pu away from the trenched area, and we also provide evidence of elevated Pu concentrations in shallow subsurface layers above the trenched area. The distribution of ^{241}Am is similar to Pu, and this is attributed to the similar chemistry of these actinides and the likely in-situ generation of ^{241}Am from its parent ^{241}Pu. Concentrations of ^{137}Cs are mostly low in surface soils immediately above the trenches. However, similar to the actinides, there is evidence of elevated ^{137}Cs and ^{90}Sr concentrations in shallow subsurface layers above the trenched area. While the subsurface radionuclide peaks suggest a mechanism of subsurface transport, their interpretation is complicated by the presence of soil layers added following disposals and during the subsequent years. The distribution of ^{90}Sr and ^{137}Cs at the ground surface shows some elevated levels immediately above the trenches which were filled during the final 24 months of disposal operations. This is in agreement with disposal records, which indicate that greater amounts of fission products were disposed in this period. The surface distribution of $^{239+240}$Pu is also consistent with the disposal documents. Although there is extensive evidence of a mobile tritium plume in groundwater, migration of the other radionuclides by this pathway is limited. The data highlight the importance of taking into account multiple pathways for the mobilisation of key radioactive contaminants at legacy waste trench sites.

1. Introduction

The common approaches to managing low-level radioactive wastes in the decades following World War II often involved disposing wastes into shallow trenches. Numerous trench facilities have been reported in various countries, with examples including Maxey Flats in the United States (Cleveland and Rees, 1981), and the Chalk River site in Canada (Jackson and Inch, 1983). These sites were typically used for wastes arising from nuclear facility operations, but they were sometimes established following accidents, such as the trenches in the vicinity of the Chernobyl area which contain debris from the 1986 reactor accident (Bugai et al., 2012; Levchuk et al., 2012). These trench disposal sites can be collectively referred to as 'legacy trench sites'.

Sites for such facilities were not necessarily carefully chosen or well-characterised before disposals commenced, with operational convenience often a deciding factor. The construction of these trenches varied; some were engineered but others were simply excavations into the soil, often without adequate waste packaging. In many countries, the standards of regulatory supervision during the period of operation were inadequate and were subsequently increased following investigations of the performance of these types of disposal sites (INEL, 1994; Takamura et al., 1984).

Such legacy trench sites often share characteristics which make them particularly problematic from a management viewpoint, such as a lack of clear ownership and responsibility, insufficient regulatory control, and missing information on site history, site layout and disposal

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processes. The radionuclide inventories are generally poorly quantified, and the plumes may be uncharacterised in terms of both the nature and extent of contamination. Furthermore, trench sites differ in terms of their contents, design, operation and geologic setting and are typically highly heterogeneous, containing a wide variety of wastes. For these reasons detailed studies of such sites are warranted and the International Atomic Energy Agency (IAEA) has promoted an international effort to compare legacy sites in various countries to evaluate the types of management strategies that may be justified and the factors which may differentiate sites requiring remediation (IAEA, 2017).

Legacy trench sites are located across the world, including some countries without major nuclear programs or power generation capabilities. One example is the Little Forest Legacy Site (LFLS) on the southern periphery of Sydney, Australia, where low-level radioactive wastes were disposed by burial in closely spaced unlined trenches between 1960 and 1968. A previous paper discussed the distribution of plutonium in surface soils and groundwater at this site, and described a process known as the 'bathtub' effect which has been implicated in the mobilisation of actinides (Payne et al., 2013), and occurs when the former trenches become filled with water during intense rainfall events. Furthermore, a groundwater plume of tritium has been delineated (Hughes et al., 2011). In recent years, the Australian Nuclear Science and Technology Organisation (ANSTO) has been implementing a detailed scientific study of the status of the LFLS (Payne, 2012) with the ultimate aim being to evaluate possible management options for the site.

In this paper we discuss recently collected data for a broad suite of radionuclides ($^{239+240}$Pu, ^{241}Am, ^{137}Cs, ^{90}Sr, ^{60}Co and ^{3}H) in both soil and water samples, and investigate various potential migration mechanisms, including groundwater transport and the bathtub effect. We will review the status of knowledge regarding the distributions of radionuclides at the site and their relationship to historic information such as disposal records. We will also discuss the mechanisms of radionuclide migration and the tendency of each radionuclide to be transported by particular pathways.

2. Site description

The LFLS is situated near the city of Sydney in eastern Australia. It occupies a site on the northern edge of the 1.6 km radius buffer zone around the location of the former HIFAR Research Reactor at Lucas Heights, which operated between 1958 and 2007. The trenches were filled sequentially from 1960 until the cessation of disposal operations in 1968. The waste disposed in the trenches included contaminated items from the operation of the Lucas Heights research facility, chemicals, disused equipment, laboratory trash, sludge drums, and beryllium wastes (Payne, 2012).

One of the main factors for site selection was the proximity to the facility which generated the majority of the waste disposed in the trenches, however the specific location was chosen so that the trenches were excavated in the surface layers within a lens of shale (Payne, 2015). This was considered to be more favourable for radionuclide retention than the prevalent sandstone geology of the area. The trenches were nominally 25 m long, 0.6 m wide and 3 m deep, and spaced about 2.7 m apart. The climate of the site is described as warm temperate with average annual rainfall of 1013 mm and average evaporation of 1600 mm (Hughes et al., 2011). However rainfall is strongly episodic and the site is subject to very intense precipitation over short periods. A detailed account of the geology and groundwater hydrogeochemistry has described the general characteristics of the site (Cendón et al., 2015) and provides a context for the radionuclide distribution data presented herein. The geophysics and geology of the site was discussed by Hankin (2012).

There are substantial records of the disposal operations, however, in common with many other disposal sites of this period, the records are incomplete. The data for different radionuclides were not individually recorded, and interpreting the records presents major challenges. For example, disposals of ^{90}Sr and ^{137}Cs were not differentiated and were generally reported among a category described as 'mixed fission products'. Furthermore, the radionuclide content of some disposed items was not evaluated or recorded, including numerous drums of sludge from a waste treatment plant (Payne, 2012). Despite the shortcomings of the waste characterisation and disposal records, they provide some valuable information for the interpretation of environmental radionuclide measurements.

There have been significant changes in land use around the LFLS since the cessation of radioactive waste disposal operations, with major industrial waste, sewage disposal and municipal landfill facilities being operated in the vicinity. The LFLS has been fenced and secured, and has been the focus of ongoing maintenance, surveillance and monitoring (Payne, 2012). The regular environmental monitoring, which has focused on the groundwater pathway, has not detected any off-site radionuclide migration, other than tritium, from the wastes buried at the site. Following the detection of surface contamination above the trenches in the early 1980s, the trenched area was covered with an additional layer of soil (AAEC, 1985). Further additions of soil have occurred on other occasions to address subsidence of the site, although detailed records are not available. Monitoring of gamma radiation within the trench area, at a height 1 m above ground-level, consistently indicates no significant difference from background levels (Hoffmann et al., 2008).

3. Methodology

3.1. Groundwater sampling and analysis

Groundwater samples were obtained from wells recently installed during the current research project, in addition to wells in the existing sampling network installed for regular environmental monitoring (Hoffmann et al., 2008). The locations of the sampling wells discussed in this paper are shown in Fig. 1a. The wells typically sample groundwater from depths between 2 m and 4 m, although some older wells were not screened to sample water from specific depths. Full details of the construction and screened intervals of the sampling wells were reported by Hankin (2012).

An additional water sampling point was installed into a former waste trench when the soil cover above a trench collapsed in August 2011 (shown as 'trench-sampler' in Fig. 1a). This sample location provides water extracted from within the trench, which contains significant amounts of actinides including plutonium (Payne et al., 2013). The vicinity around the trench sampler appears to be a source of surface actinide contamination from previous bathtubbing events. Fig. 1b shows an expanded view of the trenched area with surface contour lines indicated.

Groundwater samples were obtained using plastic tubing and a peristaltic pump. Field parameters were measured using an in-line flow cell with a portable multi-probe system (YSI 556 MPS) with pH/ORP, conductivity/temperature and DO sensors. The sampling wells were pumped until field parameters were constant prior to sample collection. All waste-water generated during sampling was collected, removed from the site and disposed of through ANSTO's waste management system. Unless unfiltered samples were of interest, water samples were filtered using an inline filter (Waterra 0.45 µm pore-size). Samples were collected and acidified to $\mathrm{pH} < 2$ with nitric acid for subsequent radionuclide measurement. For groundwater samples, 10 L of water was generally collected for radiochemical analysis, however for more active samples 100–500 mL was sufficient.

All radiochemical analyses were undertaken using standard separation and measurement techniques, followed by alpha spectrometry (actinides), gamma spectrometry (^{137}Cs, ^{60}Co) or liquid scintillation counting (^{3}H and ^{90}Sr), with purification and counting techniques as described in Harrison et al. (2011) and Hughes et al. (2011).

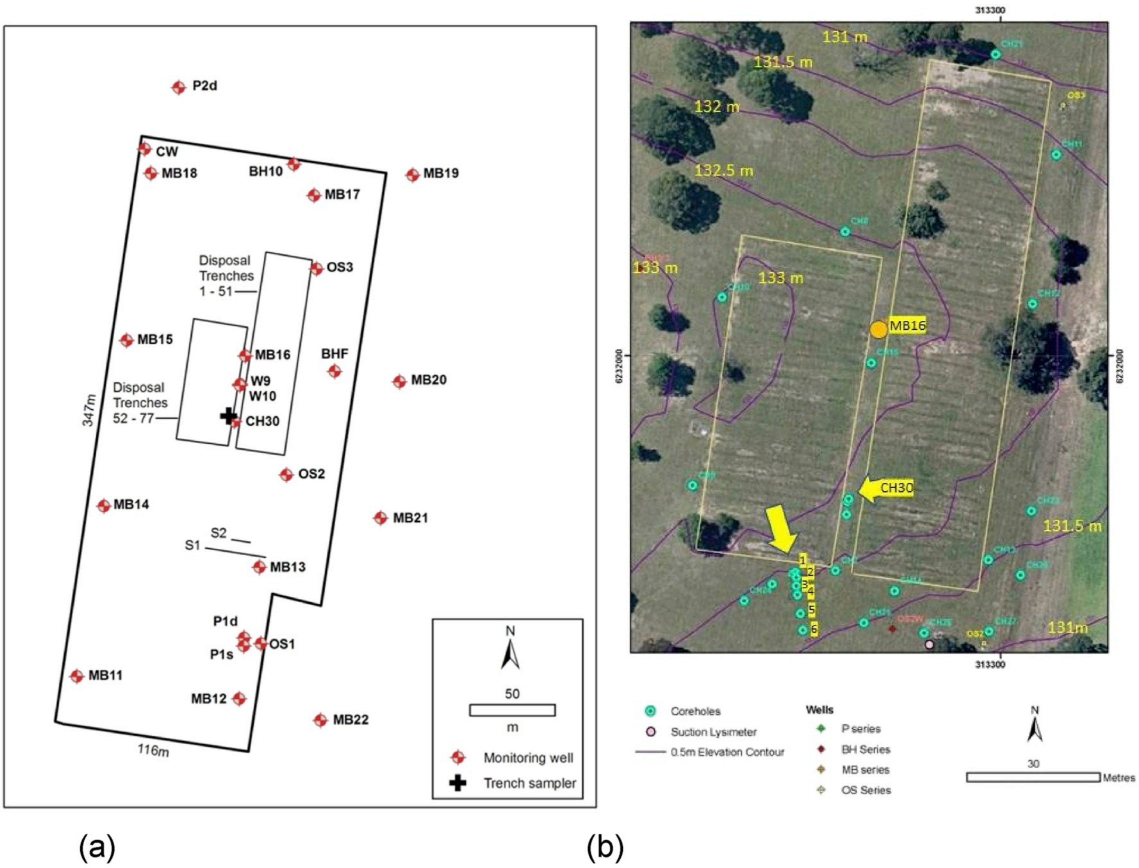


Fig. 1. (a) Map of the Little Forest Legacy Site (LFLS) showing water sampling points and (b) Aerial view of the trench area showing coreholes and surface contours. The rectangles show the two trenched areas, which are separated by a narrow gap a few metres across. The arrow on the left of panel (b) shows the location of a line of coreholes (CH1 to CH6) adjacent to the trenches for which depth profiles are presented in this paper. CH1a was an additional hole drilled adjacent to CH1. The irregular shape around the site in part (a) is the perimeter fence.

3.2. Soil sampling and analysis

Surface soils were collected with a 50 mm diameter push-tube sampler, which obtains a sample of approximately 500 g from the top 10 cm of soil. The sampling points discussed in this paper include the locations described previously (Payne et al., 2013), with numerous additional samples being obtained. The final set of samples included locations in circles at a radius of 1 m and 3 m from the trench sampler, and samples taken at 6 m and 10 m distance in the downhill direction from the trench sampler. Samples were obtained along two transects away from the trenched area down the hill-slope towards an ephemeral creek, which receives surface run-off from the site. Numerous samples were taken above the trenched area, particularly focusing on sites near the ends of the trenches. Further samples were obtained from sites within the perimeter of the fenced area (Fig. 1a), near two additional disposal trenches S1 and S2 located away from the main trenched area, and in the vicinity of a (now demolished) storage building, located at the south-west corner of the site, which was used for the storage of more active samples while their short-lived activity content decayed. Some waste drums were also stored in this area prior to burial. (The locations of all surface soil samples are shown in the Figures in the results section).

Drilling above or into the trenches has not been performed, due to the risk of disturbing buried objects and waste sources. Therefore, soil profile samples were obtained for a line of coreholes CH1, CH1a, CH2, CH3, CH4,CH5 and CH6 immediately adjacent to the trenched area, as well as CH30 which is located between the two sets of trenches (Fig. 1b).

The CH1-CH6 transect is shown in cross section in Fig. 2. At the left of the diagram is the most southerly of the trenches in the western block (Trench 52). The purpose of sampling from this transect was to assess the extent of subsurface migration from the trenches into the surrounding near-surface layers, particularly the former topsoil and surface cover layers (Fig. 2). Corehole CH30 is located between the trenches as shown in cross section (Fig. 3). The depth and spatial extent of the cover layer above the trenched area are not precisely known.

As described in detail by Hankin (2012), these coreholes were drilled using direct-push coring which enabled the retrieval of intact cores. However, this technique was only possible until the depth of ‘practical refusal’, which was variable depending on the specific location. Where it was required to extend the holes deeper, they were subsequently over-drilled with a 125 mm solid flight auger. During this process additional samples were obtained by retrieving soils and fragmented materials from the auger flight.

The corehole and surface soil samples were oven dried at 60 °C to constant weight. Corehole samples were more consolidated than surface soils and were gently disaggregated with a mortar and pestle. All soil samples were sieved through a 2 mm mesh, riffled and split, and ground in a ring mill using a carbon steel grinding head. Subsamples of the soils were leached using boiling aqua-regia reflux prior to radiochemical separation and alpha source preparation (Harrison et al., 2011). Following the sample pretreatment, radiochemical analyses were undertaken using the standard separation and measurement techniques described above.

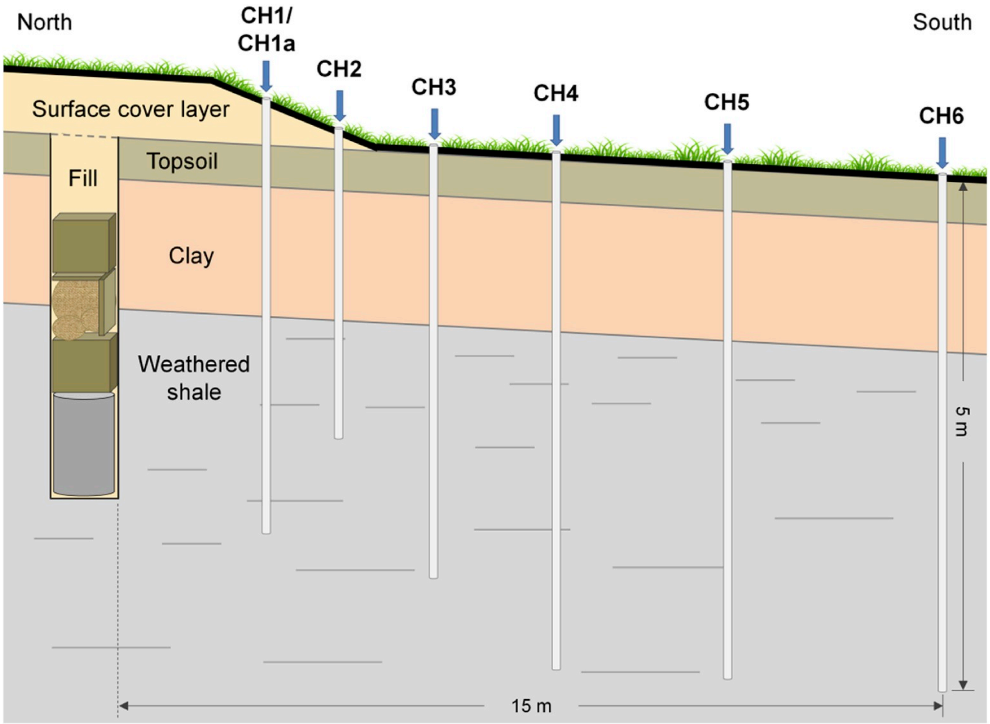


Fig. 2. Schematic cross section of the transect from CH1 to CH6. This is a transect moving away from the former trenches with CH6 located approximately 15 m from the nearest trench. The depth and spatial extent of the surface cover layer above the trenches is not exactly known. There is a slight slope from left (north) to right (south). CH1 and CH1a are adjacent to each other.

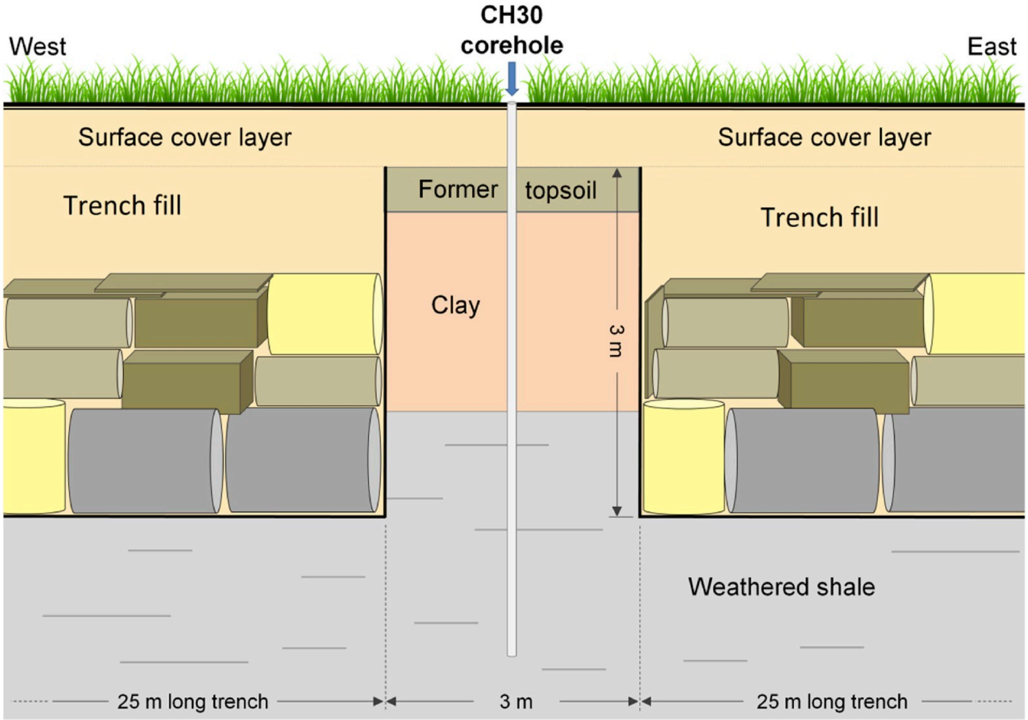


Fig. 3. Schematic cross section of the vicinity of corehole CH30. This is located on the trenched area between the two sets of trenches. The trenches are orientated from west to east and only the ends of the trenches are shown in the above Figure. Note that the exact depths of the cover layers are not precisely known (see text).

4. Results and discussion

4.1. Concentrations of radionuclides in surface soils

4.1.1. Actinides ($^{239+240}$Pu, ^{241}Am) in surface soils

The distribution of $^{239+240}$Pu in surface soils, measured by alpha spectrometry, is shown in Fig. 4. Note that alpha-spectrometry is not able to resolve these two Pu isotopes and they are counted together and referred to as $^{239+240}$Pu. The data supplement our previous results for a smaller data set (Payne et al., 2013).

The Pu data confirm that there is a significant source of Pu contamination in the trenched area, with the most highly elevated concentrations observed at the southern end of the second (western) set of trenches. This location is near the trench sampler, where significant levels of both Pu and Am have been detected in samples of trench waters (Payne et al., 2013). The peak of Pu activity in surface soils in the vicinity of the trench sampler is several orders of magnitude above typical background concentrations in the area, attributable to fallout, which range from 0.02 Bq/Kg to 0.52 Bq/Kg (Smith et al., 2016). As will be discussed in more detail below, the Pu distribution in environmental samples at the site is consistent with the known locations of Pu disposals.

There is some evidence for contamination of Pu down the hillslope from the trenched area to the south-east, with several elevated Pu measurements in surface soils obtained just off the trenched area in this direction. There is a possibility that subsurface flow through the soil layers above the trenches impacts this set of samples (shown

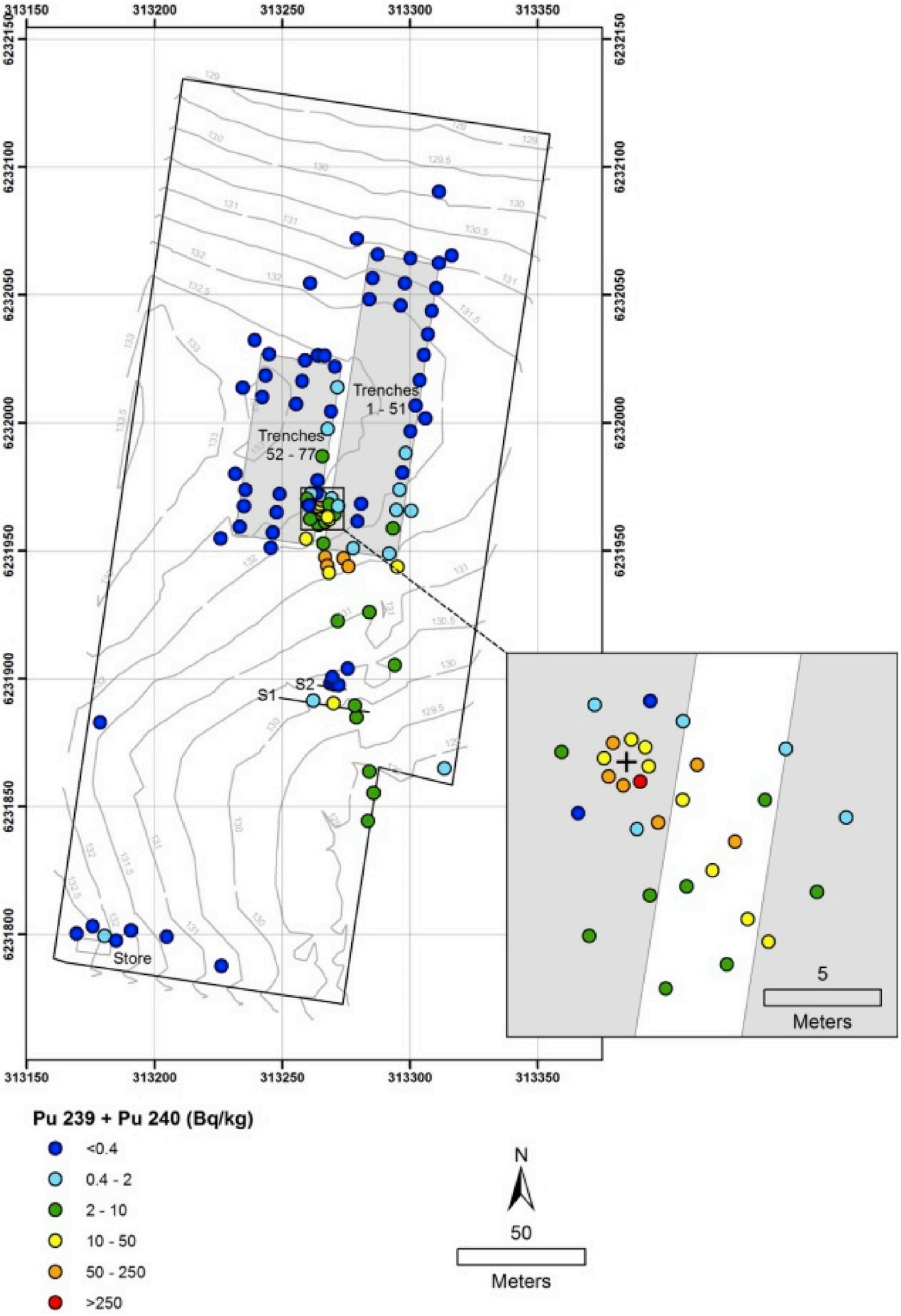


Fig. 4. Distribution of $^{239+240}$Pu in surface soils at the Little Forest site. The panel on the right hand side expands the area near the trench sampler (indicated by +). The main trench areas are shown in grey, with the two remote trenches S1 and S2 some distance away. The temporary storage building was located at the south-west of the site.

predominantly in orange in Fig. 4). This possibility is considered further below when depth profiles are discussed. However, the $^{239+240}$Pu values in the majority of samples from elsewhere on the perimeter of the trenched area are not significantly elevated, represented by deep blue in the figure (<0.4 Bq/Kg).

Another noticeable feature of the data is some samples with elevated Pu concentrations in the vicinity of trenches S1 and S2, indicating possible disposals of Pu-contaminated items in these trenches. The disposal records are not conclusive on the contents of individual trenches, however, there is evidence that a glovebox from a laboratory which handled actinides was disposed in trench S1. One sample obtained near the temporary store in the south west of the site area exhibits a slightly elevated Pu concentration, however this area generally exhibits low Pu levels (radioactive items were stored both in and around this building during the 1960s).

The distribution of ^{241}Am in surface soils at LFLS (Fig. 5) is similar to that of $^{239+240}$Pu, with the highest ^{241}Am concentrations being in the vicinity of the trench sampler. Similar to $^{239+240}$Pu, the ^{241}Am distribution exhibits a slight increase in the vicinity of trenches S1 and S2, but is relatively low (<0.5 Bq/Kg) in most samples taken around the periphery of the trenched area, except for the sample locations immediately to the south-east adjoining the trenched area.

Although the disposal of ^{241}Am was rarely reported at LFLS during the years of operation, it was probably present in all the samples which were heavily contaminated with plutonium, due it its ubiquitous presence in plutonium derived from irradiation of fuel in a reactor. It is likely that the inventory of ^{241}Am was derived from the ^{241}Am present in the disposed plutonium at the time of disposal, significantly supplemented by decay of ^{241}Pu in the years since disposal. A similar origin of ^{241}Am (i.e. derived from ^{241}Pu) has been postulated at the Hanford site, where the

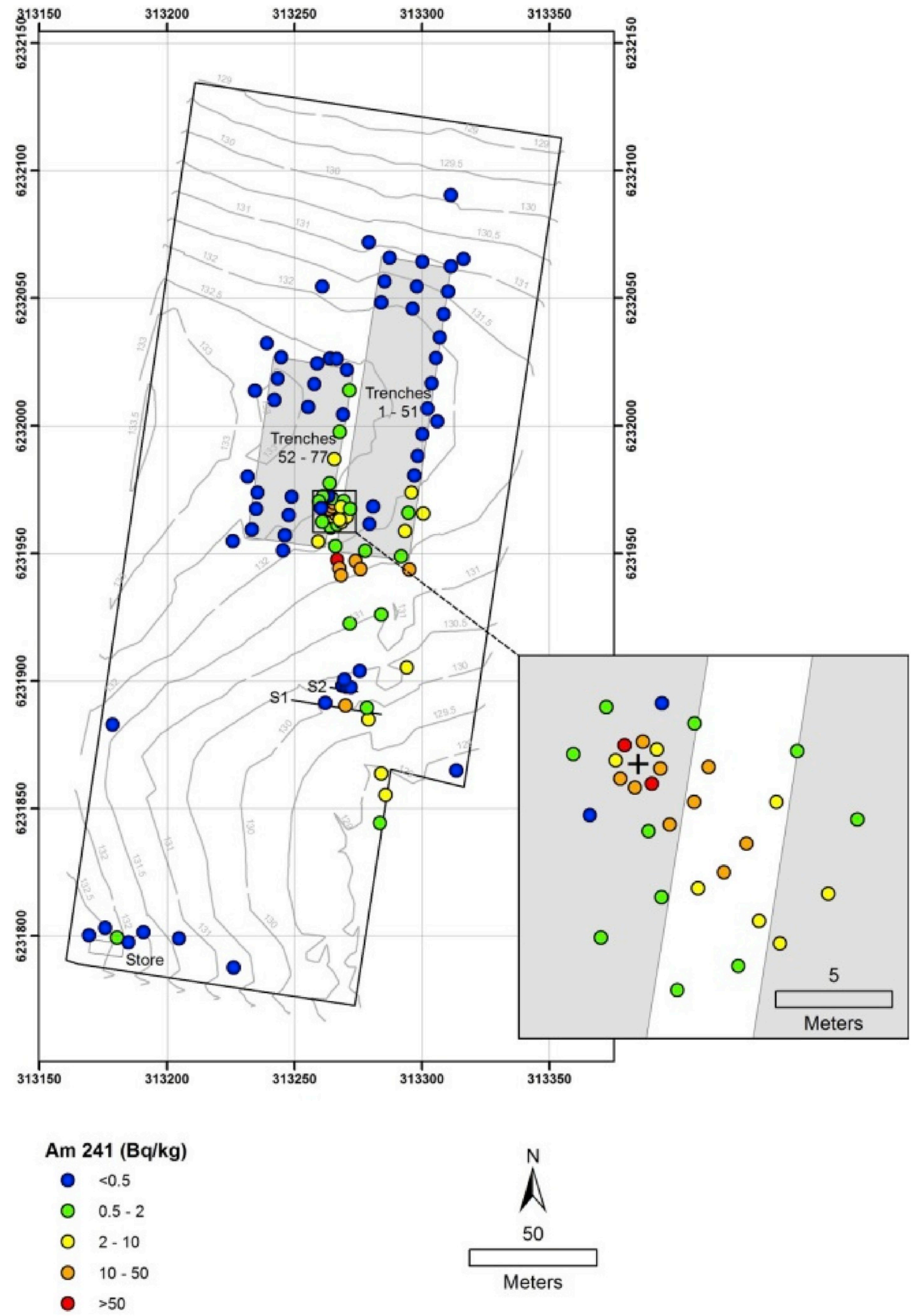


Fig. 5. Distribution of ^{241}Am in surface soils at the Little Forest site.

disposed activity of ^{241}Pu was reported to be in excess of that of ^{239}Pu and ^{240}Pu combined, even after several decades had elapsed since disposal (Cantrell and Felmy, 2012). The similarity of ^{241}Am and $^{239+240}$Pu distributions at LFLS may also be related to the chemistry of these actinides, particularly when they share the +3 oxidation state. The oxidation state and speciation of Am and Pu in trench water at the LFLS site has been investigated in detail (Ikeda-Ohno et al., 2014), with thermodynamic calculations indicating a trivalent oxidation equilibrium state for both actinides in the LFLS trench water.

The environmental mobility of Pu is influenced by its ability to simultaneously occupy multiple oxidation states, be strongly complexed with natural and co-disposed organic compounds, as well as its propensity to interact with colloids and form intrinsic colloids at higher concentrations ((Choppin, 2007; Kersting, 2013)). The mobilisation of Pu from the LFLS trenches appears to be related to complex interactions between the influx of oxidising groundwater during intense rainfall events followed by extended period of more reducing conditions, during which microbial activity affects the distribution of both actinides and other redox sensitive elements such as iron (Vázquez-Campos et al., 2017). The mechanism limiting the down-hill mobilisation of Pu and Am has not been completely resolved, but it is likely to be due to the formation of strongly sorbing Fe(III) oxides in the vicinity of the LFLS legacy trenches (Kinsela et al., 2016) as well as the substantial affinity for Pu and Am binding to sediment surfaces (Emerson et al., 2019; Lujanienė et al., 2012).

4.1.2. Fission products (^{90}Sr and ^{137}Cs) in surface soils

The distribution of ^{137}Cs in the shallow surface soil samples at the LFLS is shown in Fig. 6. The values are all quite low and are comparable to, or possibly below, fallout values for surface soils (from nuclear

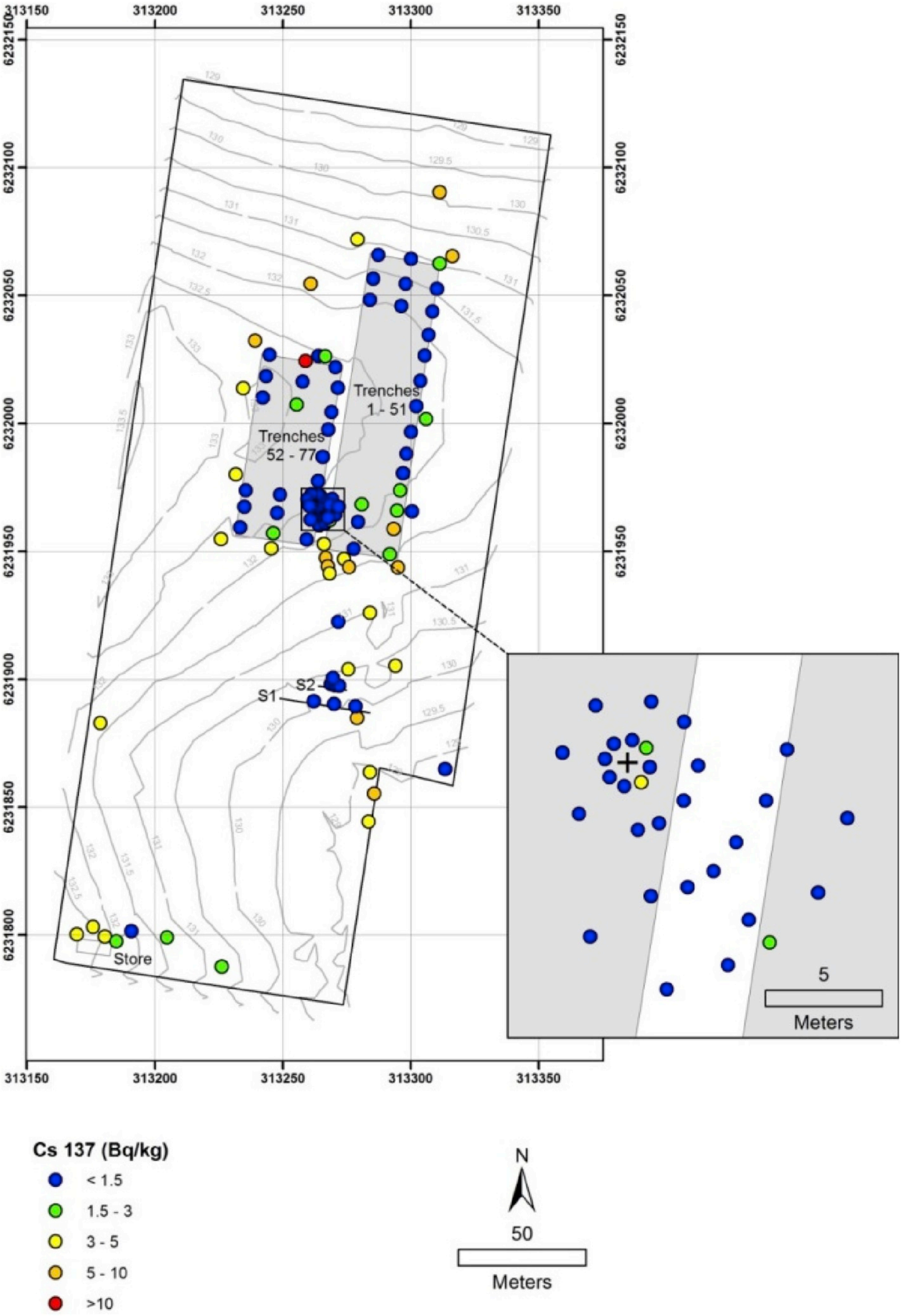


Fig. 6. Distribution of ^{137}Cs in surface soils at the Little Forest site.

weapons testing), which in this area might be expected to be similar to those reported by Smith et al. (2016) for the Sydney Basin (in the range 0.6–26.1 Bq/Kg). As such, there is little evidence for contamination of the soil surface by ^{137}Cs. However, a close examination of the data shows that the ^{137}Cs activities above the trenched area are generally lower than those away from the trenches. The ^{137}Cs activities in the vast majority of samples from the trenched area are <1.5 Bq/Kg, indicating that the level of ^{137}Cs is comparable to detection limits, and lower than fallout (Fig. 6). It should be recognised that the majority of fallout in Australia was derived from nuclear weapons tests during the 1950s and 1960s. This means that any fallout which deposited on the LFLS site has probably been disturbed by human activities at the site, and in particular, the fallout signal has been removed from the trenched area, which was excavated and covered in soil both during and subsequent to disposal operations. It is likely that the fill dirt added both during and after operations was depleted in fallout, given that the fill materials would have been obtained from below the immediate surface layer of soil. There is a single elevated measurement of ^{137}Cs in the vicinity of the final trench filled (Trench 77), which can be explained on the basis of the disposal records (discussed below).

The distribution of ^{90}Sr in surface soils (Fig. 7) resembles that of ^{137}Cs. Similar to ^{137}Cs, the trenched area is not generally elevated in ^{90}Sr, but there are some higher measurements in the vicinity of the final trenches filled. Also, ^{90}Sr appears to be slightly elevated near the trench-sampler (inset to Fig. 7). The elevated concentrations of ^{90}Sr are more numerous than was the case for ^{137}Cs, hence it appears likely that ^{90}Sr is more mobile at the LFLS site than ^{137}Cs. The activity of ^{90}Sr in fallout has been estimated as being approximately 60% of that recorded for ^{137}Cs (Mietelski, 2010). On this basis (and considering the published ^{137}Cs data of Smith et al. (2016)), fallout ^{90}Sr concentrations in these soils of

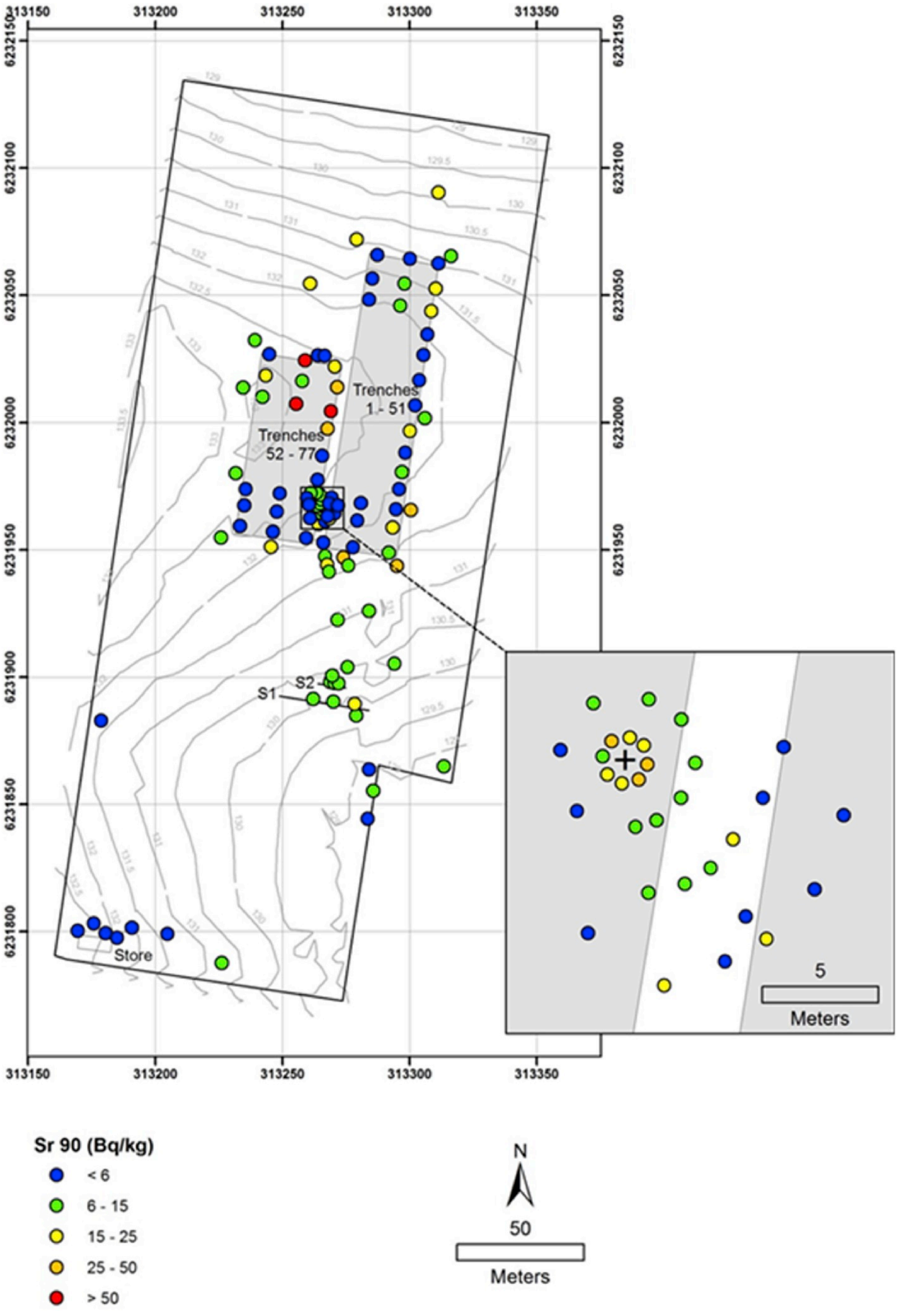


Fig. 7. Distribution of ^{90}Sr in surface soils at the Little Forest site.

up to 15–20 Bq/Kg are possible. However, the estimate of ^{90}Sr based on the most similar samples, in terms of particle size range and geographic proximity reported in the study of Smith et al. (2016), would probably be below 4 Bq/kg. Furthermore the ^{90}Sr signal from fallout may be significantly decreased by leaching from the soil. These considerations make it difficult to ascertain which points depicted on Fig. 7 might be considered as possibly impacted by the disposed waste. It appears that the only significant cluster of elevated ^{90}Sr concentrations is in the surface soils above the final few trenches filled. However, there is also a set of elevated readings around the edges of the trenched area, which may be related to flow through the surface layers above the trenches (discussed below).

4.1.3. *Correlation of radionuclides in surface soils with disposal records*

As has been demonstrated in the preceding sections there is strong evidence for $^{239+240}$Pu and ^{241}Am contamination of surface soils in the vicinity of the 'trench sampler', which is located in the southern end of the second set of trenches, among trenches which were filled in 1966 (Fig. 4). Similarly there are a few surface soil samples with elevated ^{90}Sr, mostly near the final trenches (disposals stopped in 1968), with one of these samples also exhibiting a higher ^{137}Cs signal (Figs. 6 and 7).

As part of our investigations into the LFLS, a significant effort has been expended into compiling the disposal records (Payne, 2012). Fig. 8a shows the quantities of plutonium disposed (grams per year) and of reported 'Group II' activity (Group II was a designation that included mixed fission products, such as ^{90}Sr and ^{137}Cs).

Disposals of plutonium peaked in the later part of 1965 and 1966. As shown in Fig. 8b, the eastern set of trenches was filled from 1960 to 1965, with the remaining trenches being subsequently filled from 1965 until 1968. The records therefore place the main disposals of Pu in the southern most part of the western set of trenches. Although ^{241}Am disposals were not specifically recorded, they would be expected to have a similar distribution to that of $^{239+240}$Pu. Therefore, the plutonium (and ^{241}Am) detections in surface soils at the southern end of the second (western) set of trenches are consistent with the yearly Pu disposal records and the order of filling the trenches. There is also a possibility that accidents and leakages during disposal operations may have led to radionuclide contamination of the surface soil layers at the site. For example, it was reported that numerous waste drums stored at the site in 1963 were deteriorating and susceptible to leakage during handling (Bonhote, 1964). Many of these drums were eventually buried in the trenches.

The records for disposals of Group II activity show a different pattern to the actinides. There was a large peak in disposals at the end of disposal operations Fig. 8a). This corresponds to the burial of a large number of higher activity items which had been stored (in the storage building) while the short-lived radionuclides decayed. Furthermore, it is believed that disposals were accelerated in this time due to pressure caused by the upcoming cessation of disposals. The history of disposals is therefore reflected by the distributions of ^{90}Sr and ^{137}Cs in LFLS surface soils. In summary, it can be stated that the local elevations of plutonium, as well as of ^{90}Sr and ^{137}Cs in surface soils, can be explained on the basis of the historical disposal records. In addition, leakages and accidents may also have contributed to the observed distributions of radionuclides at the site.

4.2. *Depth profiles of radionuclides*

4.2.1. *Depth profiles of actinides ($^{239+240}$Pu and ^{241}Am)*

The Pu depth profiles were obtained for the CH30 location within the trenched area and a closely spaced transect just off the trenched area (CH1 to CH6). The locations and depths of these coreholes are shown in cross section in Figs. 2 and 3. The corehole CH30 is located between the trenches in close proximity to the 'trench sampler', an area where surface soils exhibit elevated Pu concentrations (Fig. 4). This area has been substantially impacted by the addition of excavated trench soils during the operational period and also by subsequent addition of surface layers. These layers were added to address surface contamination by radionuclides in the 1980s (AAEC, 1985) and more recently to level the site and address subsidence.

The distribution of Pu at CH30 shows two strongly delineated peaks in the surface layers, the first at 20–30 cm and a second at 70–90 cm below the current ground surface (Fig. 9). The peaks may represent subsurface movement through more permeable layers above the trench, either through (now buried) shallow surface soils that pre-date the trenches, or through the disturbed layers added during and subsequent to the disposal period (see Fig. 2). Another possible explanation is that the peaks are preserved remnants of previous surface bathtubbing events which have subsequently been buried by additional surface layers. This is more likely in the case of the shallower of these two layers, as the addition of 20–30 cm of soil since the bathtubbing event in the early 1980's would be consistent with documentary evidence (AAEC, 1985; Hankin, 2012). The deeper of the two contaminated layers (at 70–90 cm) may be the result of ongoing migration through the disturbed surface layers which resulted from the disposal operations.

The Pu profile (Fig. 9) shows that the major peaks of Pu contamination in the surface layers of CH30 greatly exceed any Pu detections at deeper depths in the same corehole. However, the CH30 data also show a broad elevation in Pu concentration across all depth ranges below 2 m. This broad peak is more than two orders of magnitude below the activity of the $^{239+240}$Pu peak at the shallower depths. Furthermore, when interpreting this data, it needs to be taken into account that the samples from depths below 2 m were retrieved from augering, rather than being intact samples obtained by direct push ('practical refusal' in this borehole occurred at 2 m depth). Augering is not a controlled method of sample retrieval and it therefore appears likely that the auger became contaminated by the elevated Pu in the shallower soils during insertion and removal from the hole. Thus, we interpret the apparent diffuse subsurface peak at depths greater than 2 m in CH30 with caution, due to the method of sample collection.

The results for the CH1 to CH6 transect samples (Fig. 9) showed a much lower level of $^{239+240}$Pu contamination than CH30. However, the CH1a borehole, which is within a few metres of the nearest trench, shows Pu peaks within the top 100 cm at similar depths to the peaks in CH30. This provides evidence of some subsurface movement of Pu beyond the trenched area in this direction. Although the depth of the trenches is from approximately 100 cm–300 cm below the original ground surface, there was no conclusive evidence of migration of elevated levels of $^{239+240}$Pu at these depths beyond CH1a. This suggests that the subsurface groundwater pathway, which is responsible for significant transport of tritium (discussed further below) is not a major pathway for plutonium movement away from the trenched area. In the shallow layers (<20 cm), the $^{239+240}$Pu content of the cores of the CH1 – CH6 transect exhibits a clear decrease with depth. Although this resembles a fallout profile at an undisturbed location, $^{239+240}$Pu concentrations are over an order of magnitude higher than fallout in the Sydney basin and the site was subject to excavation and disposal operations across the maximum fallout time period (1950s and 1960s). Therefore, it is likely that at this location the surface soils were contaminated either during operations or following dispersion of contamination from the nearby trenched area.

A comparison of the CH1a profile and that of CH30 shows there is no broad Pu peak in CH1a below 220 cm in depth. This is partly attributed to the fact that the entire CH1a core was obtained by direct push coring, and augering was not used to obtain any samples in this profile. As a result, the potential contamination from shallower depths was avoided in CH1a. Additionally, a comparison of CH1 and CH1a profiles shows that the Pu peaks which are evident at shallow depths in CH1a are not well delineated in CH1. This is attributed to the lower sampling frequency used in CH1 which apparently resulted in the narrow peaks being missed when CH1 was sampled. This example shows the importance of high frequency sampling to obtain accurate depth profiles. It

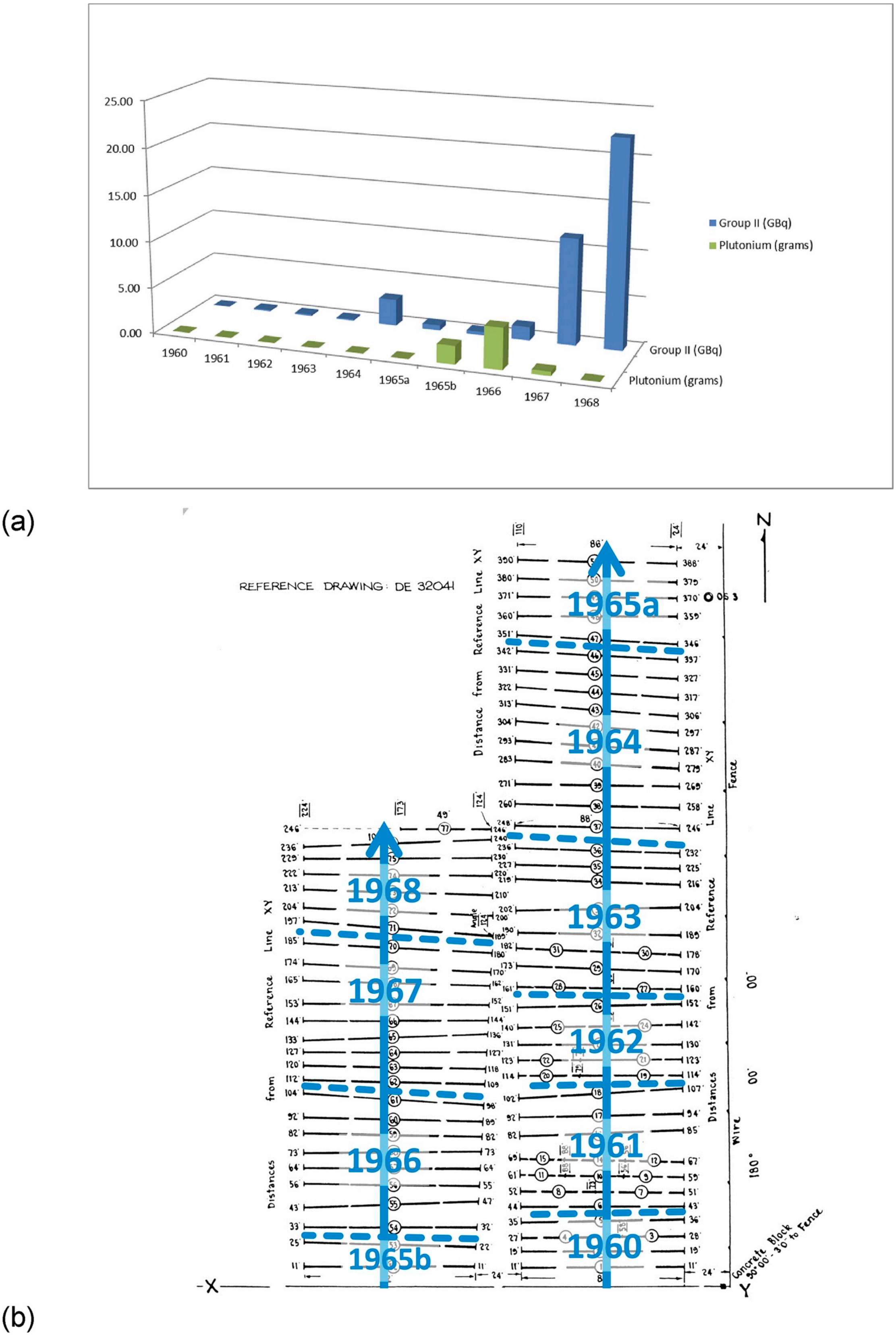


Fig. 8. (a) Disposal information of plutonium and group II activity (which included mixed fission products) at the Little Forest site for each year from 1960 to 1968 (b) Order of filling of trenches and location of trenches filled in each year, superimposed on on original AAEC plan of the trenched area. Note the activity disposed during 1965 is split into two parts (1965a and 1965b), which were disposed in different sections of the trenched area. The locations of the elevated $^{239+240}$Pu, ^{137}Cs and ^{90}Sr in soil samples show a good correlation with the records.

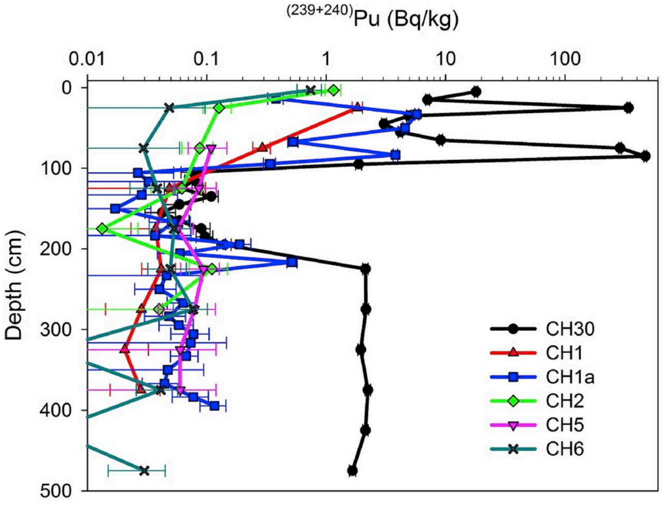


Fig. 9. Depth profiles of $^{239+240}$Pu in corehole soils.

may also reflect that the distribution of Pu in the soils is highly heterogeneous and there are slight differences between the CH1 and CH1a profiles despite their close proximity.

The depth profiles for ^{241}Am (Fig. 10) in the transect profile (CH1 to CH6) are very similar to those of $^{239+240}$Pu. The CH30 profile shows marked ^{241}Am peaks in similar depth ranges to the Pu peaks. The peaks of ^{241}Am in the shallow depths of CH30 considerably exceed ^{241}Am both at deeper levels in CH30 as well as in the CH1 to CH6 transect. However, there is some evidence of migration of ^{241}Am at the level of the base of the trenches (~320 cm) in the CH1 sample, and particularly, in the nearby CH1a profile. This slight elevation of ^{241}Am does not extend as far as CH6. Similar to the Pu, there is a broad peak in the deeper samples from CH30 (apparent in Fig. 9) which is attributable to the method of obtaining the deeper cores from this location.

The similarity in the distribution of ^{241}Am and $^{239+240}$Pu appears to be due to the similar source terms and migration mechanisms of the two actinides, and the fact that the ^{241}Am is probably partly formed in situ from β-decay of ^{241}Pu. ^{241}Am was likely present in the waste at the time of disposal due to radioactive decay of the parent radionuclide, ^{241}Pu. Although never quantified nor recorded in disposal records, ^{241}Pu may well be ubiquitous in disposed Pu as a result of neutron capture on ^{240}Pu during irradiation.

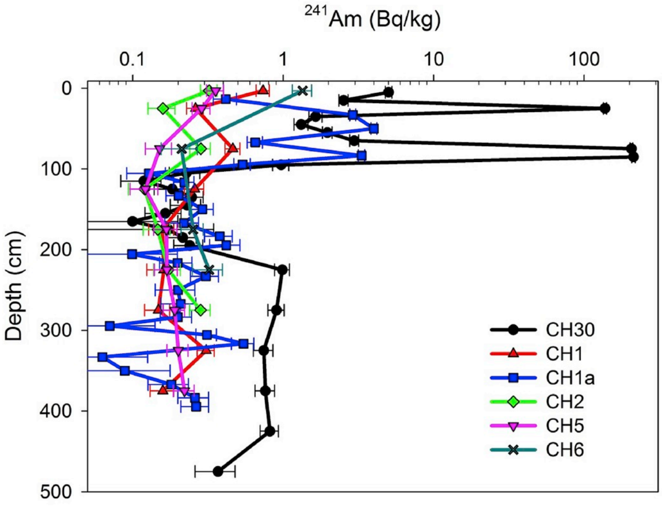


Fig. 10. Depth profiles of ^{241} Am in corehole soils.

4.2.2. *Depth profiles of fission products (^{137}Cs and ^{90}Sr)*

The depth profiles of ^{137}Cs (Fig. 11a) have some minor differences to those of the actinides previously discussed. While the CH30 profile for ^{137}Cs shows a similar distinct peak at 70–90 cm (as was found for the actinides), the corresponding peak at 20–30 cm is almost absent. This may be because there has been little mobility of ^{137}Cs into this section of the cover layers. However, it may simply also reflect that the peaks in the CH30 profile result from specific buried sources which are hydrologically upgradient. It is unlikely that any contaminant plumes in these layers would be uniform in direction and homogeneous, so the absence of a ^{137}Cs peak at this depth in CH30 does not necessarily imply there is no ^{137}Cs at this depth elsewhere within the site.

Some of the CH1 to CH6 transect samples, located just away from the trenched area, show a small peak at the surface. This is most likely caused by ^{137}Cs particles being washed out by a surface transport pathway from the nearby trenched area (although it cannot be totally excluded as being related to fallout). Surface transport is also postulated as a mechanism of movement of actinides down the hillslope, responsible for the dispersion of actinides across the ground surface. However, in general the amount of ^{137}Cs surface contamination is relatively small.

There are two peaks of ^{90}Sr in the depth profile of CH30 (Fig. 11b), with the upper peak being greater. These peaks are somewhat broader than was the case for ^{137}Cs and the other radionuclides. Thus, we can

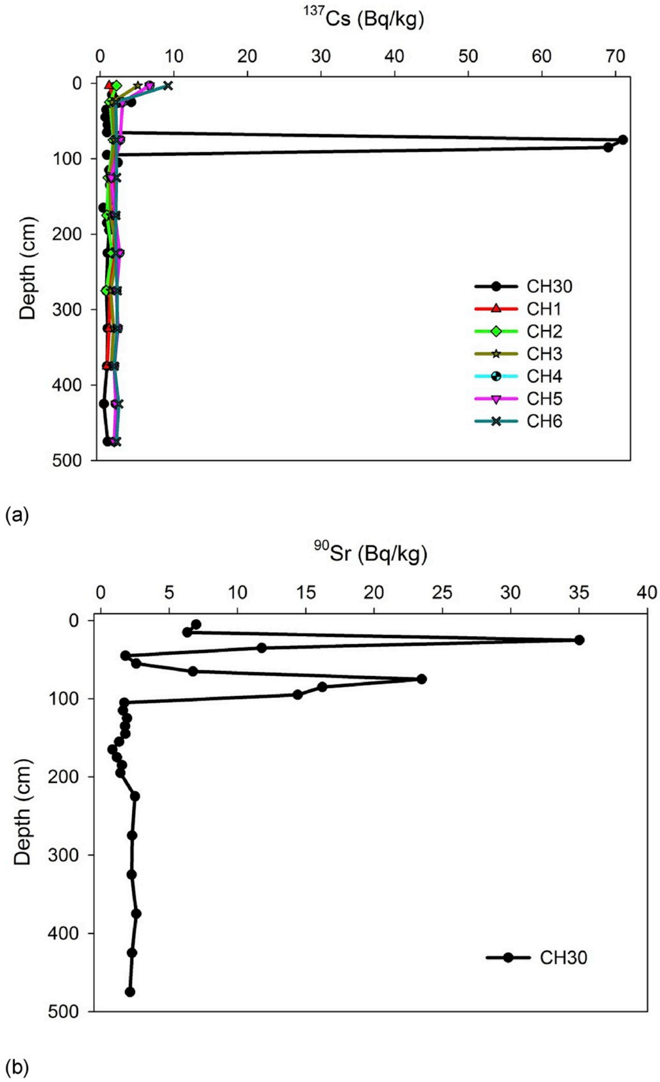


Fig. 11. Depth profiles of fission products (a) ^{137}Cs and (b) ^{90}Sr (for CH30 only).

conclude that ^{90}Sr is comparatively mobile in the near surface layers and that the peak has spread vertically to a greater extent. All measurements of ^{90}Sr in the CH1 to CH6 transect were below minimum detectable activity (MDA) values, however due to the less sensitive measurement methods for this radionuclide, it is not possible to rule out mobility of ^{90}Sr through this transect.

In general it would be expected that ^{90}Sr would be more mobile than ^{137}Cs as the distribution coefficients for ^{90}Sr retention by soils are typically much lower than corresponding values for ^{137}Cs (Sheppard and Thibault, 1990; Twining et al., 2004), The immobility of ^{137}Cs in this environment would be consistent with the clay dominated shale layers and soils in this area, which are favourable for Cs sorption (Cornell, 1993).

The radionuclide data for the fission products in the uppermost cover layers, within approximately 1 m from the ground surface, provide additional evidence that these layers facilitate the subsurface transport of radionuclides (which is also demonstrated by the subsurface peaks of the actinides $^{239+240}$Pu and ^{241}Am). Movement of radionuclides through these layers, with emergence at the ground surface at the edge of these added layers (see Fig. 2) is also likely to be responsible for the elevated local concentrations of $^{239+240}$Pu, ^{241}Am and ^{90}Sr just beyond the south-eastern corner of the trenched area (Figs. 2, 3 and 5, respectively).

The core logs reported in Hankin (2012) mention the presence of fill materials in coreholes above the trenched area, however, it is difficult to delineate the depth and spatial extent of the added surface layers at LFLS. This is because these layers are mainly composed of fill excavated from the trenches and then placed above the wastes, which means that they are chemically and mineralogically similar to the underlying layers. The radionuclide peaks found in the upper layers may be attributable to pathways arising from the physical processes of excavation, re-emplacement, ground levelling and possibly compaction. In addition, the uppermost layer is also probably impacted by the addition of soil from local sources applied during top-dressing of the site in the 1980s and subsequently to address subsidence and bathtubbing events.

4.2.3. Depth profile of ^{60}Co

Significant amounts of the short lived activation product ^{60}Co (half-life 5.3 years) were disposed during the operational period of the Little Forest site. This was partly due to the irradiation of samples using ^{60}Co sources which were operated at the AAEC site in the 1960s. These activities led to ^{60}Co being one of the major contributors of the original inventory of activity. In the years immediately following the cessation of disposals, it was one of the most commonly detected radionuclides in environmental samples, with detections in groundwater close to the trenches and also in vegetation growing above the trenches during the 1970s (AAEC, 1985). Several decades have now elapsed since disposal operations, which means that the ^{60}Co has decayed to a fraction of its original activity (<0.2% over 50 years), and is rarely detected in environmental samples. There is a subsurface peak of ^{60}Co in the CH30 profile (Fig. 12), which occurs at a deeper level than for the radionuclides discussed above (^{241}Am, $^{239+230}$Pu, ^{137}Cs, ^{90}Sr). This depth is more likely to be more continuously saturated, than shallower depths which only become saturated during elevated rainfall periods, when the water level in the trenches rises higher. The reasons for the difference between the depth profiles of ^{60}Co and the other radionuclides are unclear but may be related to different trench water chemistry (more reducing conditions) when the water levels are lower, which is known to impact the mobility of radionuclides in the system (Ikeda-Ohno et al., 2014).

The ^{60}Co migration mechanism therefore appears to be more similar in character to that of tritium than the other radionuclides considered here. The migration of cobalt can be enhanced by organic complexes (Collins and Kinsela, 2010) which have also been implicated in Pu mobility at LFLS (Rowling et al., 2017). Whilst the reasons for the differences between ^{60}Co and the other radionuclides are not entirely clear, the distinctive behaviour of this radionuclide shows how different pathways may be dominant for the various radionuclides. The different behaviour of ^{60}Co underlines the complexity of fully understanding the system at LFLS and the necessity to evaluate multiple possible migration pathways.

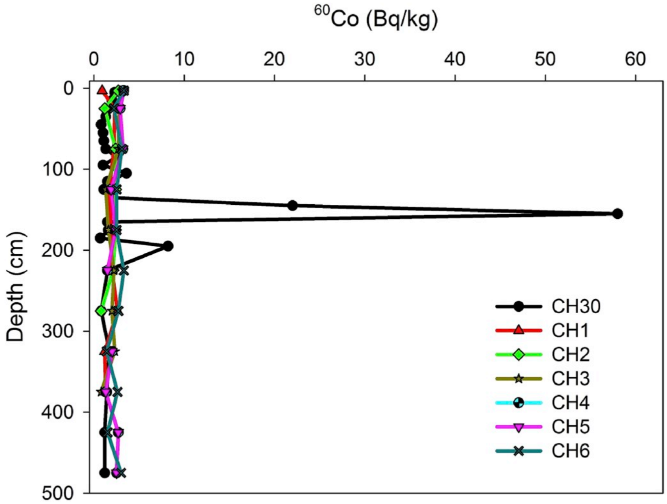


Fig. 12. Depth profile of ^{60}Co showing evidence of horizontal movement around 150 cm depth.

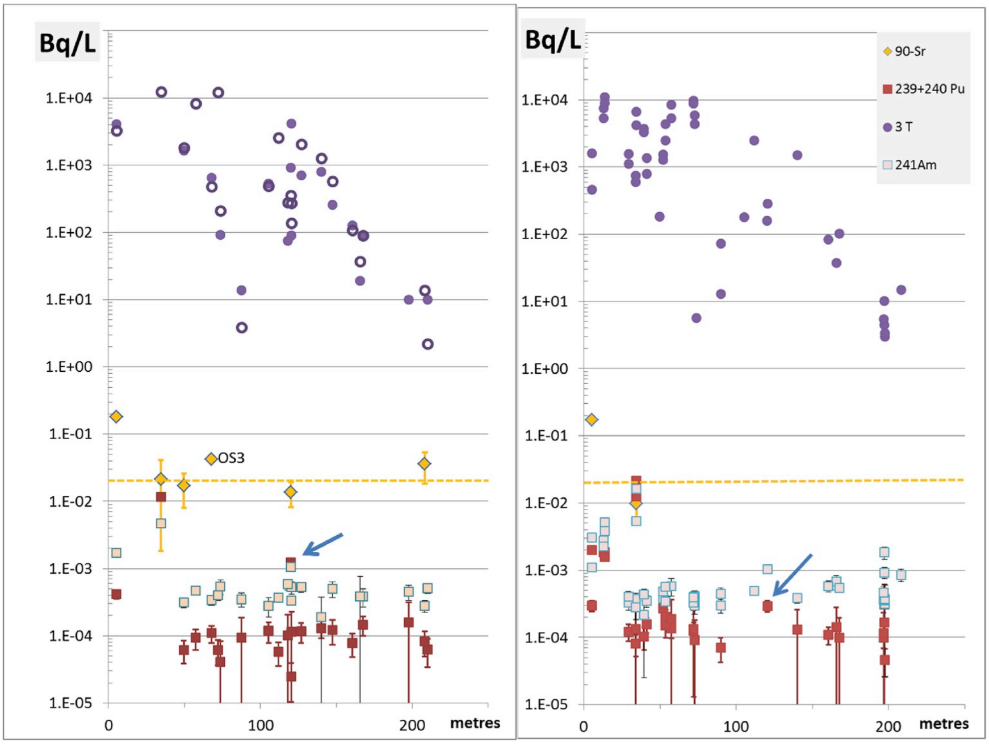
4.3. Radionuclides in groundwater

The measured concentrations of radionuclides (other than tritium) in LFLS subsurface water samples are typically very low, in most cases bordering on detection limits, even for samples gathered within or near the trenched areas. This creates difficulties in generating contour diagrams of the data (as was done for the tritium data by Hughes et al. (2011)) because the data interpolations do not converge. However, despite the inherent challenges in measurement and interpretation of very low values, it is important to establish whether or not there is significant migration of radionuclides in LFLS groundwater.

To assess the groundwater data for $^{239+240}$Pu, ^{241}Am, ^{90}Sr and ^{137}Cs, we therefore compared their distributions to the tritium data. As discussed by Hughes et al. (2011), the contour mapping of the tritium data results in approximately concentric circular contours originating from the trenched area. Although there are some preferential directions, the location of the site on a local high point causes tritium to generally spread out radially rather than in a linear fashion (ground level contours are shown on Fig. 1b). Therefore, to a first approximation, the trenched area can be considered to be a point source (although a relatively large and heterogeneous one). This provides a method for presenting the data for the other radionuclides and comparing them with the tritium data.

Thus, we selected a ‘zero’ (origin) of distance at the centre of the trenched area, located at the coordinate position 313275 m (east) and 6232000 m (north). This position is very close to the location of borehole MB16 (Fig. 1), which consistently has given amongst the highest readings of radionuclides in groundwaters at the site. We then determined the distance of all other sampling wells from this origin, using borehole coordinates mapped by GPS. This method provided simple plots of all data as a function of distance from the approximate centre of the trenched area (as shown in Fig. 13).

We first consider the results for tritium, as the characteristics of tritium movement at the site are well understood. Fig. 13a shows the average of groundwater tritium data (for the period 2003 to 2008) which were tabulated in Hughes et al. (2011) arranged by distance from the origin (multiple measurements were made at each sampling point). This has been supplemented with a second tritium data set from 2009, which was taken in conjunction with measurements of the other radionuclides shown in the figure. Similarly, Fig. 13b shows tritium data,



(a)

(b)

Fig. 13. Data for tritium (^{3}H), fission products and actinides in LFLS groundwaters. (a) Averaged tritium data for the period 2003 to 2008 (solid circles), plotted with a groundwater sample set for 2009 (^{90}Sr, $^{239+240}$Pu, ^{241}Am and ^{3}H). The ^{3}H for the 2009 data set is indicated by open circles. (b) Measurements for 2010 and 2011 data sets, in which all samples were measured for ^{90}Sr, $^{239+240}$Pu, ^{137}Cs, ^{241}Am and ^{3}H. All measurements are expressed as activities at the time of sampling. The measurements for $^{239+240}$Pu at MB13 are arrowed (see text). ^{137}Cs concentrations are not shown as all samples returned values below MDA (0.01 Bq/L). The MDA for ^{90}Sr was sample specific and depended on chemical recovery. The dashed orange line shows a typical MDA for ^{90}Sr in LFLS water samples. In a few samples ^{90}Sr could be quantified (e.g. well OS3) and these data points are shown. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

and that for other radionuclides, for samples obtained during sampling campaigns in 2010 and 2011. It can be seen that in all these cases, there is clearly an elevation of tritium near to the origin with a gradual decline moving away from this point. The trenched area extends for approximately 50 m from the distance origin and therefore all the data points for distances below 50 m were obtained between, or close to, the trenches (e.g. boreholes MB15, W9, W10 or CH30, see Fig. 1a). From approximately 50 m–200 m away from the centre of the trenched area on this distance scale, the tritium activity concentrations decrease in a fairly systematic fashion, as is expected when moving away from a point source of contamination, with all data sets showing consistent tritium trends.

The simplified approach used in Fig. 13 is helpful in interpreting the data for the other radionuclides. Whilst a few samples close to the distance origin show slightly elevated levels of the actinides (^{241}Am, $^{239+240}$Pu), the concentrations of these radionuclides outside the trenched area (>50 m from the distance origin) show virtually no spatial trends. In particular, the regular decrease exhibited in tritium concentrations is absent. It should be noted that many of the measurements are close to detection limits.

While many measurements were at, or barely above detection limits, it would be reasonable to question why any of the data points showed any measurable amounts of actinides. However, it needs to be taken into account that the LFLS is a contaminated site with multiple potential radionuclide sources. Furthermore, some contaminated samples are many orders of magnitude above background and well in excess of detection limits. This means that multiple sources may contribute to the data at any given location, including surface water flow from the main trenched area, particles transported during heavy rainfall events, contamination (e.g. accidents) which may have occurred during disposal operations, and/or airborne particles, as well as fallout. An illustrative example is the Pu data obtained for MB13 groundwater (shown with arrows in Fig. 13a and b). This location is adjacent to one of the isolated disposal trenches (S2), see Fig. 1a, which constitute an additional radionuclide source separate to the main trenched area. Hence, there is a plausible reason for the elevated Pu in groundwater at this location (a similar elevation in soil Pu levels near the S1 and S2 trenches was discussed above). All the remaining Pu data points outside the trenched area exhibit no apparent distance-related trends.

All ^{137}Cs measurements were below MDA for ^{137}Cs (~0.01 Bq/L) and ^{60}Co was not detected in any groundwater samples. Therefore, data for these radionuclides are not shown in Fig. 13.

^{90}Sr could only be definitively measured in a few water samples from within the trenched area (Fig. 13), and was elevated in samples from well OS3, located just away from the edge of the trenched area (Fig. 1a). Although there were a small number of other detections outside the trenched areas, these ^{90}Sr levels were close to MDA limits for each sample. The MDA for individual samples depends on factors such as sample recovery and the sample mass available for analysis, and as such is not constant. The typical individual sample ^{90}Sr MDA is indicated by a dashed orange line in Fig. 13. As can be seen, there is no strong evidence that ^{90}Sr measurements beyond the trenched area were above this value.

The low levels of $^{239+240}$Pu in LFLS groundwaters outside the trenched area (typically around 0.1 mBq/L, see Fig. 13) are significantly below those found in the water sampling point in the former waste trench (Payne et al., 2013), which were approximately 5 orders of magnitude higher (11.1–12.6 Bq/L in filtered water). They are also much lower than the groundwater levels of $^{239+240}$Pu reported in the close vicinity of some Chernobyl trenches containing Pu contamination, which were in the range 1–360 mBq/kg (Levchuk et al., 2012). The greater migration reported in the latter case is attributable to the sandy environment of the Chernobyl trenches, the greater source term, and the smaller scale of the sampled region. Similarly the ^{90}Sr groundwater contamination in the vicinity of the Chernobyl trenches (in the range 4–4500 Bq/kg) is many orders of magnitude higher than the ^{90}Sr measured in any LFLS samples, including those taken from the sampling points between the trenches.

The primary conclusion from Fig. 13 is that, unlike tritium, the groundwater pathway has not been a significant migration mechanism over decadal timescales for $^{239+240}$Pu, ^{137}Cs, ^{241}Am, or, with somewhat less certainty, ^{90}Sr. For these radionuclides, much higher levels of activity are found in the soil profiles and surface soils.

5. Conclusions

The information gained from the detailed studies at LFLS provides insights into the environmental mechanisms leading to their migration at the site and will help inform the choice of remediation options. The distributions of radionuclides at LFLS are a combination of their source terms and their mobility, and are also impacted by events during the history of the site, including past additions of surface layers. The work described in this paper demonstrates some of the challenges associated with environmental sampling of contaminated sites, in particular the very large range of contaminant concentrations observed (which raises issues of detection limits and potential cross contamination). In addition, limitations of available methods of site characterisation and sample collection can complicate the interpretation of the data (for example, the Pu data for the CH30 profile exhibited in Fig. 9). Nevertheless we have assembled robust and comprehensive data sets which enable significant conclusions to be drawn.

For both ^{137}Cs and ^{90}Sr, the concentrations which have arisen beyond the trenched areas are negligible and, even where elevated by migration from the trenches, are not significantly above the range expected for environmental samples of the region due to fallout. The highest level of both radionuclides in surface soils is within an order of magnitude of fall-out values. In this respect, the data for the fission products differ significantly from the measured $^{239+240}$Pu and ^{241}Am results, where the peak concentrations were several orders of magnitude above expected fallout concentrations in surface and subsurface soils. However, significantly elevated concentrations were generally limited to the immediate vicinity of the trenched area, with a small contribution attributed to overland particle movement during rainfall events. The distributions of ^{137}Cs, ^{90}Sr, ^{241}Am and $^{239+240}$Pu in surface soils are most highly elevated in close proximity to their known disposal locations based on the preserved records. Although there has been evidence of elevated ^{60}Co in some environmental samples shortly after disposal, the ^{60}Co has mostly decayed and there is no evidence of ^{60}Co contamination apart from within the subsurface layer in the trenched area.

The existence of subsurface peaks of contamination in the cover layers above the trenches needs to be taken into account if the site is remediated, as it may add to the volume of contaminated material which would need to be re-disposed. This will depend on the level of contamination relative to limits for disposal and the remediation strategy. Further field measurements and experimental work will be required to evaluate the different migration mechanisms.

In summary, the different radionuclides at the site exhibit different patterns of mobility. Tritium is mobile through the groundwater pathway, with some evidence for limited groundwater mobility of ^{60}Co and ^{90}Sr. The actinides (^{241}Am and $^{239+240}$Pu) are dispersed by a bathtub mechanism which brings them to the surface during sufficiently intense rain events. There are also subsurface peaks of these radionuclides in the shallow cover layers above the trenches, which may be due to subsurface flow or previous bathtubbing events. Elevated levels of both ^{90}Sr and ^{137}Cs are found in these layers.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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