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Accelerator mass spectrometry measurements of ^{233}U in groundwater, soil and vegetation at a legacy radioactive waste site

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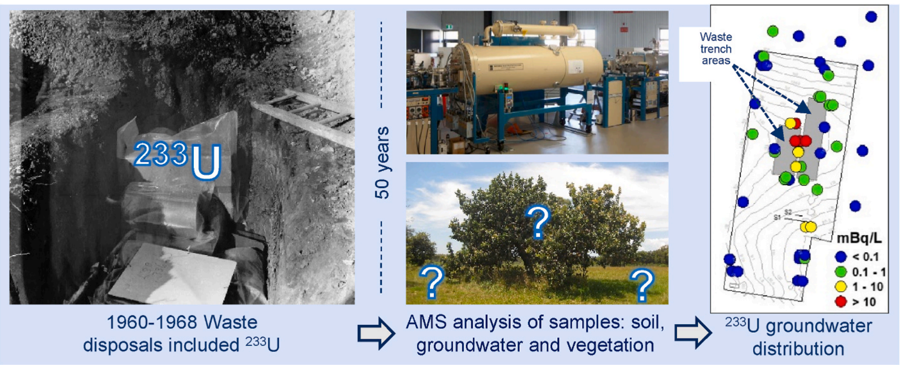
HIGHLIGHTS

- First results for ^{233}U contamination at a legacy radioactive waste site.
- ^{233}U data obtained by Accelerator techniques.
- Unusually high levels of ^{233}U in many samples, consistent with disposal records.
- ^{233}U concentrations decrease with distance from waste sources.

GRAPHICAL ABSTRACT



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ABSTRACT

Low-level radioactive wastes were disposed at the Little Forest Legacy Site (LFLS) near Sydney, Australia between 1960 and 1968. According to the disposal records, ^{233}U contributes a significant portion of the inventory of actinide activity buried in the LFLS trenches. Although the presence of ^{233}U in environmental samples from LFLS has been previously inferred from alpha-spectrometry measurements, it has been difficult to quantify because the ^{233}U and ^{234}U α-peaks are superimposed. Therefore, the amounts of ^{233}U in groundwaters, soils and vegetation from the vicinity of the LFLS were measured using accelerator mass spectrometry (AMS). The AMS results show the presence of ^{233}U in numerous environmental samples, particularly those obtained within, and in the immediate vicinity of, the trenched area. There is evidence for dispersion of ^{233}U in groundwater (possibly mobilised by co-disposed organic liquids), and the data also suggest other sources of ^{233}U contamination in addition to the trench wastes. These may include leakages and spills from waste drums as well as waste burnings, which also occurred at the site. The AMS results confirm the historic information regarding disposal of ^{233}U in the LFLS trenches. The AMS technique has been valuable to ascertain the distribution and environmental behaviour of ^{233}U at the LFLS and the results demonstrate the applicability of AMS for evaluating contamination of ^{233}U at other radioactive waste sites.

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1. Introduction

There have been a number of recent studies of the anthropogenic uranium isotope ^{233}U ($T_{1/2} = 0.159$ My) in the environment, which mostly relate to dispersed environmental contamination in pristine environments (Chamizo et al., 2022a; Hain et al., 2020; Qiao et al., 2020) or ^{233}U associated with nuclear weapons test sites (Child and Hotchkis, 2013). The source of ^{233}U has primarily been the atmospheric testing of nuclear weapons, with an estimated 13 kg of ^{233}U being released into the environment due to this global source (Chamizo et al., 2022b). However, ^{233}U has also been produced and handled at nuclear research facilities (Lin et al., 2021) and has been disposed at some low-level radioactive waste sites, with one example being the Little Forest Legacy Site (LFLS) in eastern Australia (Harrison et al., 2016). The LFLS contains wastes derived from research into nuclear power generation involving ^{233}U which was undertaken during the 1960s (AAEC, 1985; Payne, 2012). The site is on the suburban fringes of southern Sydney, and has been the subject of detailed study in recent years. The work has been aimed at evaluating the present status of the site (including any environmental or dose impacts), and developing optimal management options (Griffiths and Payne, 2018).

The wastes buried at LFLS were produced during the operations of the HIFAR reactor (High Flux Australian Research Reactor) and associated research facilities. The ^{233}U disposed at the site was derived from the Australian power reactor research program, which involved two power reactor systems, the High Temperature Gas-Cooled Reactor and the Liquid Metal Fuel Reactor. It was expected that both systems could operate efficiently using the ^{233}U/Thorium breeder cycle (AAEC, 1957; Hardy, 1999). Although the research programs into reactors fuelled by ^{233}U were abandoned after a few years, these research activities resulted in some materials contaminated with ^{233}U being disposed at the nearby LFLS. According to the accepted standards of the time, the low-level wastes were disposed into unlined trenches excavated in shallow clay-rich soil layers (Payne, 2012).

The current research at the LFLS has involved an extensive investigation of the disposal records and estimation of the radionuclide inventories (Payne et al., 2021, 2022). The only anthropogenic isotope of U mentioned in these records was ^{233}U, although it is possible that ^{236}U and ^{232}U were also disposed at the site. The study described in this paper is therefore focused on ^{233}U, which is known to have been disposed in the LFLS trenches.

The records indicate that gram quantities of ^{233}U (~5 g in total) were buried in the trenches. Although this is smaller than the mass of disposed ^{238}U (around 50 Kg), the activity of ^{233}U (~1.9 GBq) probably exceeded that of ^{238}U (~0.73 GBq), on account of the much higher specific activity of ^{233}U. The ^{233}U is therefore one of the major long-lived sources of activity at the site. In many disposed items, ^{233}U was dissolved in solvents (Payne et al., 2022), primarily tri-butyl phosphate (TBP) and di-*sec*-butyl phenyl-phosphonate (DSBPP). Both of these solvents were used for complexation during actinide separation processes (Phillips, 1967; Rowling et al., 2017). Therefore, it is important to evaluate the environmental distribution and potential mobility of ^{233}U at the LFLS.

A recent report summarised extensive studies of the distributions of various radionuclides (e.g. ^{3}H, ^{90}Sr, ^{137}Cs, $^{239+240}Pu$, ^{241}Am) at LFLS, measured using conventional radiochemical methods such as α, β and γ techniques (Payne et al., 2020). This previous report did not include ^{233}U, because ^{233}U levels in environmental samples cannot usually be measured by standard radiochemical methods. Even the relatively high levels of ^{233}U in LFLS samples have been difficult to quantify by α-spectrometry, because the α-particles are in the same energy range as ^{234}U. Although some progress has been made in quantifying ^{233}U from α-spectra (Harrison et al., 2016), this technique is not suitable for the majority of environmental samples from LFLS, due to the ubiquitous presence of ^{234}U from natural sources and trench wastes.

The sensitive technique of Accelerator Mass Spectrometry (AMS), which discriminates between uranium isotopes based on their masses, has increasingly been applied in recent years for measurements of trace actinides in the environment (Chamizo et al., 2022a, 2022b; Child and Hotchkis, 2013; Hain et al., 2020; Hotchkis et al., 2000; Qiao et al., 2020; Wallner et al., 2022). Therefore, in the present work, we used AMS to obtain definitive measurements of ^{233}U in environmental samples from LFLS. The objective was to determine the amounts and distribution of ^{233}U in these environmental samples and to establish whether these results can be related to the history of ^{233}U disposals at the site. This paper includes AMS results for ^{233}U in 101 samples from the LFLS, including 66 groundwater samples.

2. Methodology

2.1. Site description

The LFLS occupies a site in the vicinity of the former High Flux Australian Reactor (HIFAR) at Lucas Heights, on the southern fringes of Sydney in Eastern Australia. The site is approximately 400 m long and 150 m wide, with the shallow trenches being filled sequentially from 1960 until the cessation of disposal operations in 1968. There were 79 disposal trenches, each excavated to a depth of 3 m in the clay-rich surface layers. The wastes disposed in the trenches included contaminated items from the operation of the Lucas Heights research facility, chemicals, disused equipment, laboratory trash, waste drums, and beryllium wastes (Payne, 2012). In addition to the wastes emplaced in the trenches, numerous waste items were burnt at the site, including some contaminated with radioactivity (Payne et al., 2021). The site has been fenced and maintained since the disposal era and is a predominantly grassy area, with some trees in close proximity to the trenches. The research at LFLS and the measured data for various radionuclides in the surrounding environment have been described in previous papers and reports (Harrison et al., 2016; Payne et al., 2013, 2020). There have been detailed studies of the groundwater geochemistry (Cendón et al., 2015, 2022) and of the possible presence of organic contaminants in the groundwater (Rowling et al., 2017), which may impact on the mobility of radionuclides in the local subsurface.

The plan-view of LFLS (Fig. 1) shows the main features of relevance to the study of ^{233}U distributions at the site. The two main trenched areas are in the centre of the site (the eastern Trenches 1 to 51; and western Trenches 52 to 77), with two other trenches S1 and S2 located to the south of the main trenches. The Figure shows the locations of trenches 48, 55 and 67 (T48, T55 and T67), where ^{233}U is known to have been disposed (Payne et al., 2021), although it is considered likely that other trenches also contained some ^{233}U contamination. The main area where $^{239+240}Pu$ surface contamination has been previously delineated is indicated (Payne et al., 2013, 2020). Arrows show the approximate directions of subsurface tritium transport (Hughes et al., 2011). The storage hut at the south-west corner of the site was used for the decay of highly active packages until the dose-rates were acceptable for burial. A nearby area was used for the storage of sludge drums which contained a range of radionuclides and were eventually disposed in the trenches (Payne et al., 2021).

2.2. Sampling

Groundwater samples were obtained from wells installed during the current research project (Hankin, 2012), and from wells installed for regular environmental monitoring (Hoffmann et al., 2008). 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. The selection of boreholes for sampling was a balance between a comprehensive coverage of the site, and generating a reasonable number of samples relative to analytical and resourcing constraints. Full details of the construction and screened intervals of the sampling wells were reported by Hankin (2012).

Groundwater samples were obtained using methods described

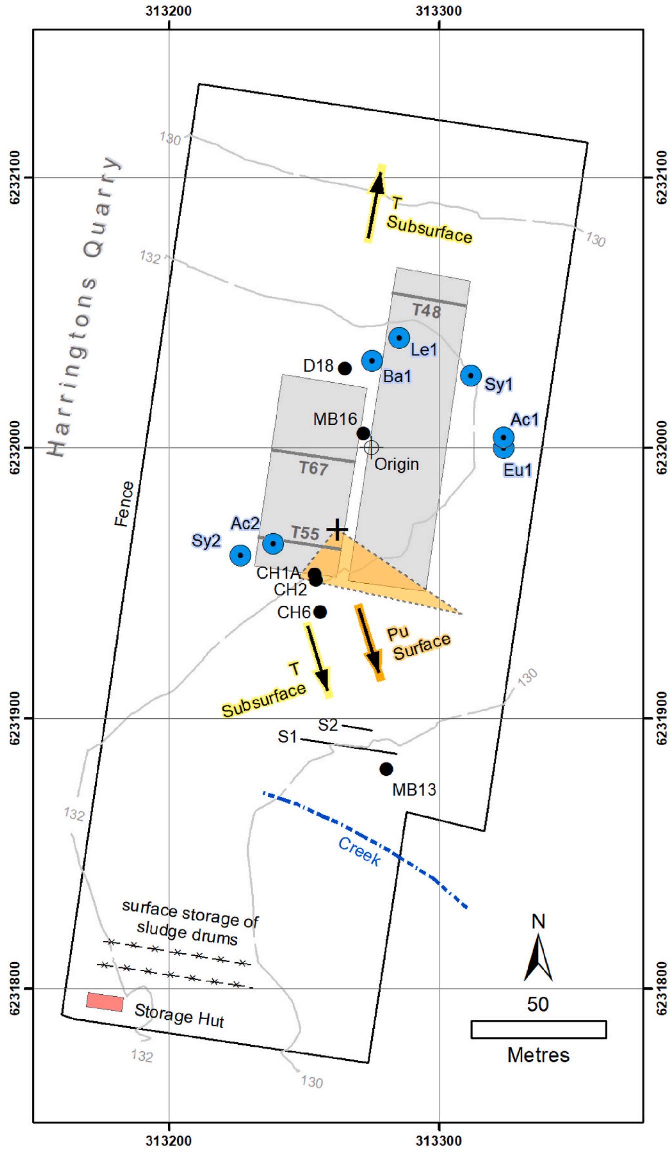

Fig. 1. Map of the LFLS showing surface contours, the two main trenched areas (grey rectangles), trenches S1 and S2, key boreholes (MB16, MB13 and D18), and coreholes (CH-1A, CH-2 and CH-6). Trenches 48, 55 and 67, in which gram quantities of ^{233}U were reportedly disposed, are indicated. The seven trees from which wood and foliage were analysed are shown as blue circles (Sy = *syncarpia*, Ba = *banksia*, Ac = *acacia*, Eu = *eucalyptus*, and Le = *leptospernum*). The main extent of Pu surface contamination and direction of surface Pu movement is indicated (orange). The dominant directions of subsurface tritium transport (Hughes et al., 2011) are shown by labelled arrows (yellow). The origin location at 313275 (east)/6232000 m (north) is indicated (see section 3.3). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

previously (Harrison et al., 2016; Payne et al., 2020) using a peristaltic pump, with field parameters measured using an in-line flow cell with a portable multi-probe system (YSI 556 MPS) with sensors for pH/ORP, conductivity/temperature and dissolved oxygen. The sampling wells were pumped until field parameters were constant prior to sample collection. Water samples were filtered using an inline filter (Waterra 0.45 μm pore-size), with unfiltered samples being collected if required. Samples were acidified to pH < 2 with nitric acid for subsequent radionuclide measurements. A groundwater volume of 10 L was generally collected for radiochemical analysis, with samples acidified with nitric acid to pH < 2 and evaporated to 500 mL on a hot-plate (for some more active samples, 100–500 mL was collected and the evaporation step was omitted). The evaporated homogenised samples were then evenly split for AMS and α-spectrometry.

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. Numerous surface soils were taken above the waste trenches, including the area (Fig. 1) known to be contaminated with radionuclides by a process known as ‘bath-tubbing’, which causes radionuclides to be brought to the surface when the former trenches become saturated during intense rainfall events (Payne et al., 2013). Additional samples were taken from the vicinity of the trenches, and from an area near a storage hut, which was used to store wastes for an initial decay period prior to disposal during operations in the 1960s.

In addition, we took depth-profile samples from some coreholes near the trenches to assess the extent of subsurface migration from the trenches into the surrounding near-surface layers. For safety reasons it was not possible to drill coreholes which intersected the trenches. The sampled coreholes (denoted CH-1A, CH-2 and CH-6) were drilled using direct-push coring which was intended to enable the retrieval of intact cores (Hankin, 2012). Samples were taken from the surface to depths of 3–5 m, depending on borehole length. The soil profiles consisted of disturbed local topsoil, as well as added soil layers above the trenches and clay-rich subsurface layers. Corehole samples were gently disaggregated as necessary with a mortar and pestle.

The corehole and surface soil samples were oven dried at 60^{o}C to constant weight. 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 (Harrison et al., 2016). This leaching procedure provides solutions of relatively simple composition suitable for radiochemical analysis and provides an estimate of the contamination associated with the soil particles (both natural and anthropogenic).

A set of 11 vegetation samples were also subjected to AMS and α-spectrometry analysis. The samples were collected from trees within and adjacent to the trenched area, including the native species *acacia longifolia*, *eucalyptus paniculate*, *syncarpia glomulifera*, *banksia serrata*, and *leptospermum polygalifolium* (Fig. 1). Wood (from tree branches) and foliage samples were dried at 40^{o}C to achieve a stable weight. Vegetation samples were ashed at 450 °C for at least 48 h using a slow ramp rate to avoid combustion, ground in a bench-grinder, then dissolved in a mixture of HNO_3 and HCl. Full details of the vegetation sampling and digestion methods are given in Wilsher (2012).

2.3. Analysis

2.3.1. Alpha spectrometry

Samples for U measurement by α spectrometry were separated and measured as described by Harrison et al. (2011). An accurately known amount (~0.2 Bq) of a tracer spike of ^{232}U (Eckert & Ziegler source 83609-657) was used to monitor chemical losses during the processing. A Canberra Alpha Analyst with 450 mm^2 passivated ion-implanted planar silicon (A450-18AM PIPS) detectors was used to count α sources using the standard counting configuration of this system (3.5–6.5 MeV energy range; 1024 channels; recoil suppression counting mode air thickness 16 μg/cm^2; source to detector distance = 0.9 cm; 48 h count time). A mixed α source (Eckert & Ziegler source 7400-SRC; 79963-121; ^{238}U, ^{234}U, ^{239}Pu and ^{241}Am) was used for energy and efficiency calibration. The $^{233+234}$U multiplet was used to determine the combined $^{233+234}$U activity, which was used to quantify the subsequent AMS results.

Unspiked samples were also analysed, to ensure that all isotopes of U were accounted for. The presence of ^{232}U in the LFLS samples was not detected, which suggested that the disposed wastes did not contain significant amounts of ^{232}U. This is consistent with the absence of ^{232}U in the LFLS disposal records.

2.3.2. AMS measurements

The AMS samples were chemically purified to isolate U according to standard methods (Harrison et al., 2016; Hotchkis et al., 2000). Prior to analysis, ^{236}U was added to the soil and water samples as an isotope dilution tracer for determination of U concentration. However, it was decided not to use this method for determination of ^{233}U concentrations due to the possible presence of ^{236}U in some wastes disposed at the LFLS. Therefore, ^{233}U/^{234}U atom ratios rather than absolute concentrations were measured by AMS, with the absolute amounts of ^{233}U determined using the total $^{233+234}$U activity concentrations from α-spectrometry. Water and vegetation samples were measured on the ANTARES accelerator at ANSTO using methods previously described in detail (Child and Hotchkis, 2013; Hotchkis et al., 2010). The vegetation samples did not have any tracer added. The soil samples were analysed using the Vega AMS system (Hotchkis et al., 2019). The ^{233}U/^{234}U atom ratios were calibrated using AMS standards of known isotopic ratio, prepared by quantitative mixing of NBL U010 standard with NBL 111-A ^{233}U spike. The ^{233}U/^{234}U atom ratios were converted to activity ratios and then ^{233}U activity concentrations were calculated using the combined $^{233+234}$U activity concentrations from α-spectrometry. In this paper, we refer to these data as ‘AMS results’, although it is noted that the analysis involves both the AMS and α-spectrometry techniques. Further details of the data processing using this method are given in Harrison et al. (2016).

3. Measured ^{233}U in groundwater

3.1. Comparison of AMS ^{233}U groundwater data and $^{233+234}$U/^{238}U ratios by α-spectrometry

The data for ^{233}U concentrations in LFLS groundwater (measured by AMS, normalised by α-spectrometry as described above) against the $^{233+234}$U/^{238}U activity ratio (directly from α-spectrometry) for sample sets collected in 3 sampling campaigns between 2008 and 2010 are shown in Fig. 2. The error estimates associated with the data take into account counting uncertainties, sample sizes and other sources of error.

The highest three ^{233}U AMS values (50–205 mBq/L) were for a single borehole (MB16) near the centre of the trenched area, with similar results obtained across all 3 sampling campaigns. Another nearby borehole (D18) was also amongst the highest ^{233}U AMS values (21.2 mBq/L). The ^{233}U values for boreholes MB16 and D18 are considerably above the AMS detection limit (estimated as ~1 μBq/L). The samples from MB16 and D18 also show highly anomalous $^{233+234}$U/^{238}U activity ratios (>10) by α-spectrometry, which are much higher than the usual range of this ratio in the LFLS groundwater samples (~1–3). The highest $^{233+234}$U/^{238}U ratios obtained by α-spectrometry correspond to the elevated ^{233}U results for the same samples by AMS (Fig. 2).

According to the disposal records, the largest disposals of ^{233}U at LFLS occurred in nearby Trench 67 in 1967 (Payne et al., 2022). This trench contained a total of 2.50 g of ^{233}U (of which at least 2.00 g was described as being a “loose powder” in disposal records), together with approximately 60 L of liquid wastes, including organic liquids. As discussed above, these liquids were used to solubilise actinides during the research which was being undertaken at the time of waste disposals at LFLS. The disposal of the organic actinide complexant Tri-butyl Phosphate (TBP) has previously been implicated in the mobilisation of plutonium at the LFLS (Rowling et al., 2017), and ^{233}U may be mobilised by a similar mechanism, particularly given its powdered form.

Excluding the most contaminated samples from MB16 and D18, the remaining samples all had somewhat similar $^{233+234}$U/^{238}U activity ratios (in the range from 1 to 3). These were not significantly different from the ^{234}U/^{238}U ratios typically found in uncontaminated water samples from the area, and for these samples the ^{233}U activity contributed a relatively small proportion of the total $^{233+234}$U activity. This means that the α-spectrometry is not generally useful for quantifying the levels of ^{233}U in LFLS samples, even using deconvolution methods (described by Harrison et al. (2016)). In contrast, numerous LFLS samples contained elevated levels of ^{233}U, which were readily measured by AMS (Fig. 2).

Because the AMS ^{233}U data for many samples significantly exceed detection limits (Fig. 2), the AMS technique is definitive in quantifying the amounts of ^{233}U present for the majority of this set of groundwater samples (n = 66). In contrast, the $^{233+234}$U/^{238}U activity ratios cover a relatively narrow range (apart from the MB16 and D18 samples) and the deconvolution of alpha spectra (using the methods described in Harrison et al. (2016)) only yields accurate ^{233}U data for the samples with the highest ^{233}U content (i.e. MB16 and D18). The data demonstrate that AMS is necessary for quantifying the ^{233}U content of most of the LFLS groundwater samples.

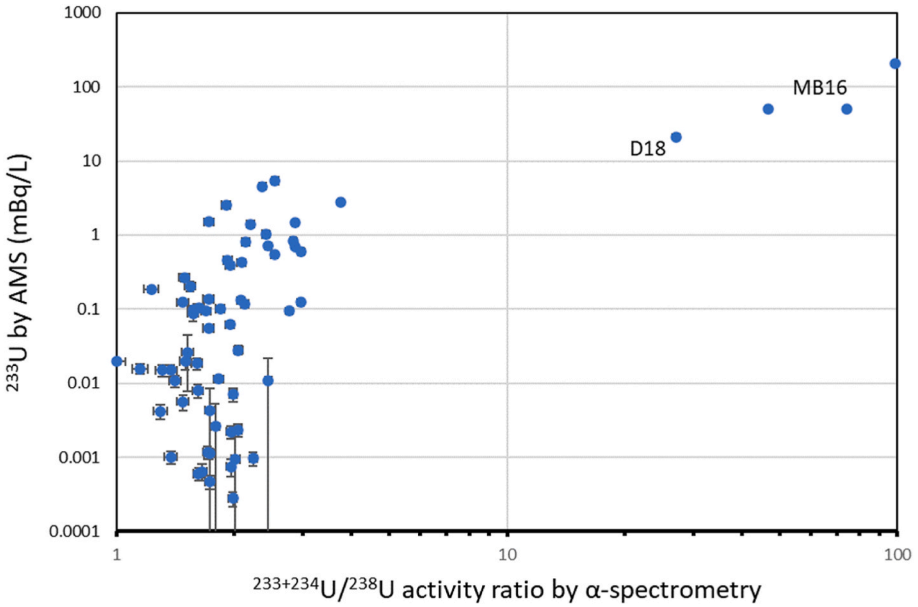

Fig. 2. ^{233}U activity concentrations in groundwater samples as measured by AMS compared to $^{233+234}$U/^{238}U activity ratio as determined by α-spectrometry. Data obtained for boreholes MB16 (3 samples) and D18 are marked. The highest ^{233}U activity was 205 mBq/L for a sample from MB16.

3.2. Spatial distributions of ^{233}U in groundwater

The spatial distributions of AMS-measured ^{233}U in groundwater samples obtained from LFLS across three sampling campaigns are shown in Fig. 3. The red symbols reflect the elevated values for boreholes MB16 and D18 (>10 mBq/L). A lower, but notable level of ^{233}U contamination (1–10 mBq/L, represented by yellow symbols) was found in several other groundwater samples taken between the main trenched areas, and from boreholes immediately adjacent to them. Samples taken from borehole MB13, which is near the two isolated trenches (S1 and S2) to the south of the main trenched areas, also fall into this category. These detections probably indicate the presence of undocumented ^{233}U disposed in these two trenches, which also are apparent sources for $^{239+240}$Pu and ^{241}Am, as indicated by groundwater data for these isotopes (Payne et al., 2020). As discussed in more detail below, there is evidence to suggest that disposals of items contaminated with ^{233}U were more widespread at LFLS than reported by the disposal records (Payne et al., 2022). However, the presence of ^{233}U in the MB13 samples could also be attributed to the movement of groundwater in this direction from the main trenched areas.

The remaining samples from around the perimeter of the site (Fig. 3) show mostly lower concentrations than in the centre of the trenched area, with possible evidence for some mobility of ^{233}U in groundwaters, particularly in the northern direction. This is similar to the direction of contaminated water movement at the site as delineated by the tritium plume (Hughes et al., 2011). There is an additional possibility that some contamination outside the trenched areas may be due to burning of waste items in unknown locations on the site, additional disposals that have not been reported, or contamination from leakages of waste packages (Payne et al., 2022). For example, waste drums were stored on the site are known to have been in a deteriorating state prior to disposal (Bonhote, 1964), and they may have leaked during storage, transport or burial.

3.3. ^{233}U in groundwater with distance

The concentrations of contaminants with distance from a postulated source can help to understand retardation process and to differentiate between point-sources and diffuse sources of contaminants (Leterme et al., 2006). In Fig. 4, the groundwater radionuclide data for ^{233}U at LFLS are presented as a function of distance from a point near the centre of the site where the groundwater levels of ^{233}U are the highest. The selected origin is near to MB16 (Fig. 1), at the coordinate position of 313275 (east) and 6232000 m (north). This location was previously used as the origin for reporting radionuclide data measured by conventional α, β and γ radiochemistry techniques (Payne et al., 2020).

The data for ^{233}U span approximately 6 orders of magnitude, and there is a general trend of decreasing ^{233}U with increasing distance from the origin in all directions. This is primarily because the LFLS is located on a slight rise, and the water generally flows in radial directions downgradient away from the site (Fig. 1). However, the data combined on this graph does not represent a single flow line, but a combination of directions. For this reason, we have indicated individual sets of points according to their compass directions (both within and beyond the fenced area), as shown by the coloured symbols on Fig. 4 and the inset diagram.

The grey symbols (diamonds) in Fig. 4 represent ^{233}U concentrations within the trenched area. In this area, the ^{233}U concentrations are considerably elevated. Even within this small area, the ^{233}U data show a sharp decrease with distance from the most contaminated area in the vicinity of MB16 and D18 (refer Fig. 1), indicating a specific source in this area, consistent with the disposal records.

We next consider the ^{233}U data from the western sector, which is generally upgradient of the trenches (data shown by red symbols on Fig. 4). There is some considerable scatter in the data, which may represent mixing effects (possibly from transient changes in groundwater direction), dilution by rainfall, and geological heterogeneity. Changes in the groundwater flow paths are expected following major rainfall events due to the ‘bathtub effect’, which causes the groundwater elevation to be raised in the trenches (Payne et al., 2013). This process brings contamination to the surface and may temporarily influence (or reverse) the hydrological gradient. This is a possible reason for elevated ^{233}U content of water samples from close to the trenches in the western sector, although local sources of contamination (such as isolated spills) may also have occurred.

The main directions of inferred water movement away from the trenches at LFLS are to the north (indicated as yellow symbols) and south (orange). The ^{233}U data in Fig. 4 clearly show this trend, with elevated data extending for ~150 m in both northward (yellow symbols) and southwards (orange) directions. The measured data are generally higher than for the western (upgradient) samples taken at a similar distance (red diamonds). There is an apparent peak in ^{233}U at borehole MB13, which is in the proximity of the two isolated trenches S1 and S2. As noted above, the presence of ^{233}U contamination here may be derived from these nearby trenches rather than by groundwater transport from the main trenched area. However, the presence of organic complexants in the groundwater may significantly increase the mobility of ^{233}U, as is postulated to occur with Pu at the site (Rowling et al., 2017).

There is limited evidence for groundwater transport of ^{233}U to the east (as shown by the blue symbols). The ^{233}U data for this sample set tend to be lower than in the other directions. It seems possible that there is either less groundwater flow or greater retardation of ^{233}U in the eastern direction. However, the data set is limited by the relatively lower number of sampling boreholes, particularly beyond the fenced area, as well as possible deficiencies in the construction of some boreholes (particularly MB19, MB20 and MB21), which may permit dilution from percolating rainwater (Cendón et al., 2015; Hughes et al., 2011).

In summary, the ^{233}U groundwater data set reported here verifies that the LFLS trenches are the major source of ^{233}U contamination at the site. The ^{233}U data are consistent with the mixing of rainfall with inflowing groundwaters, acquiring a characteristic increased signature of ^{233}U in the trenched area. The ^{233}U data show a very sharp drop off within the trenched area (grey sample set, Fig. 4), reflecting that the source of mobile ^{233}U is localised within a specific part of the trenched area. The ^{233}U data decrease consistently with increasing distance in all directions. However, outliers to this trend, particularly at MB13, indicate the possibility that localised sources of contamination (including the S1 and S2 trenches) contribute to the measured ^{233}U groundwater concentrations.

4. 4 ^{233}U in Soils

4.1. Corehole soil depth profiles

We obtained sets of ^{233}U data for depth profiles in three coreholes (CH-1A, CH-3 and CH-6) located at the southern fringe of the trenched area (Fig. 1). The data (Fig. 5) show the presence of ^{233}U in near surface samples in this region, which is thought to be impacted by radioactivity dispersed by the bath-tub effect from nearby trenches (Payne et al., 2013, 2020).

The CH-1A borehole is within a few metres of the nearest trench (Fig. 1), and exhibits a readily detected ^{233}U signal (up to 710±20 mBq/Kg) within the top 100 cm, with a similar depth profile to what was previously seen for $^{239+240}$Pu by α-spectrometry (Payne et al., 2020). Based on the ^{233}U global fallout amount of 13 Kg (Chamizo et al., 2022b), and the dimensions of the surface soil samples taken in this study (described above), we would expect that the amount of ^{233}U contributed by fallout in this sample would be below 0.1 mBq/Kg. Thus the amount of ^{233}U in this surface soil sample exceeds the estimated fallout contribution by a factor of ~10^4.

The elevation of ^{233}U in near-surface layers extends to CH 6, at a greater distance from the trenches. These results may provide evidence

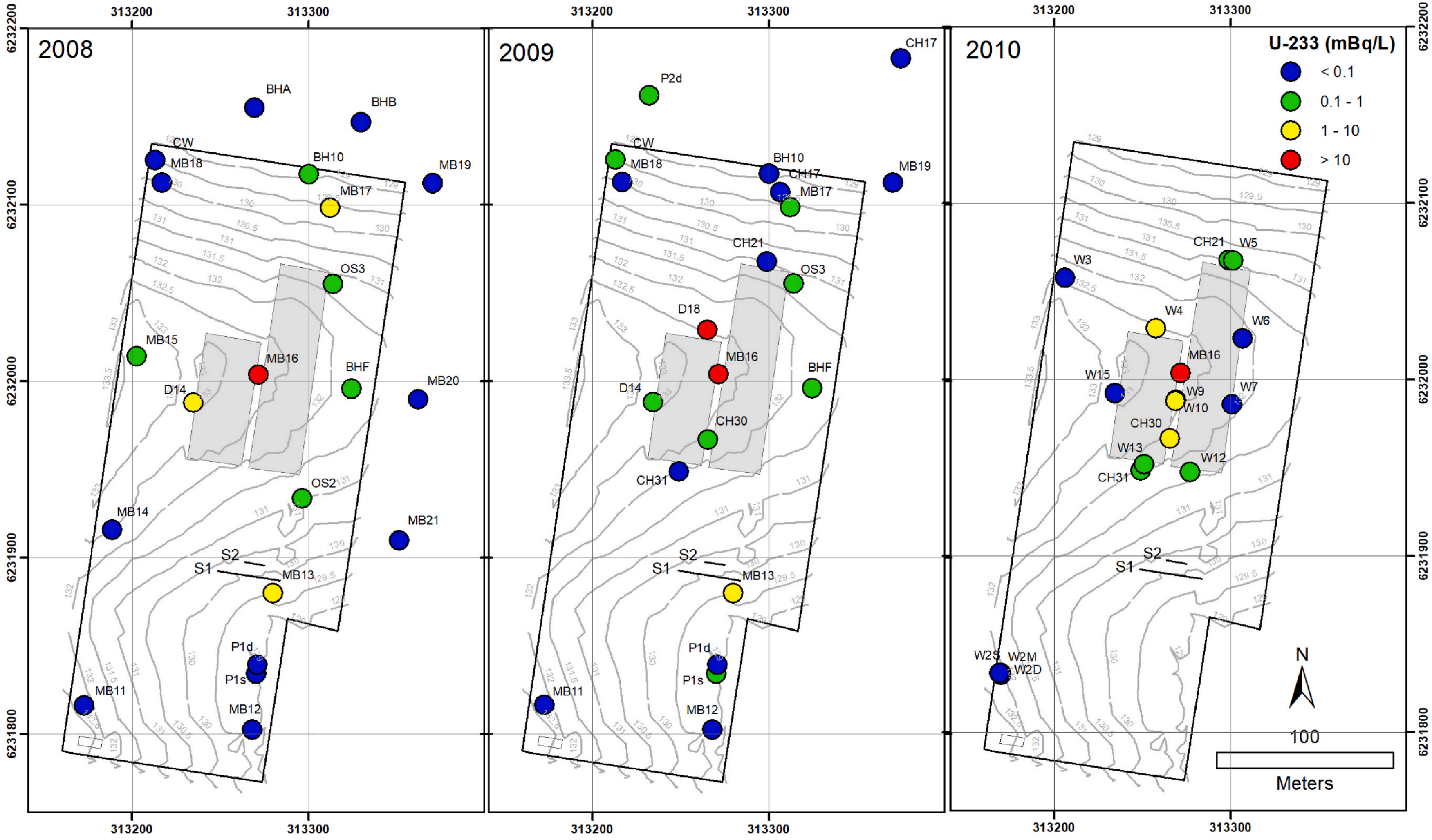

Fig. 3. The distribution of ^{233}U in groundwaters measured for samples obtained in three field groundwater sampling programs between 2008 and 2010. The two main trenched areas are the grey rectangles, with two isolated trenches S1 and S2 to the south of the main trenched areas (see Fig. 1).

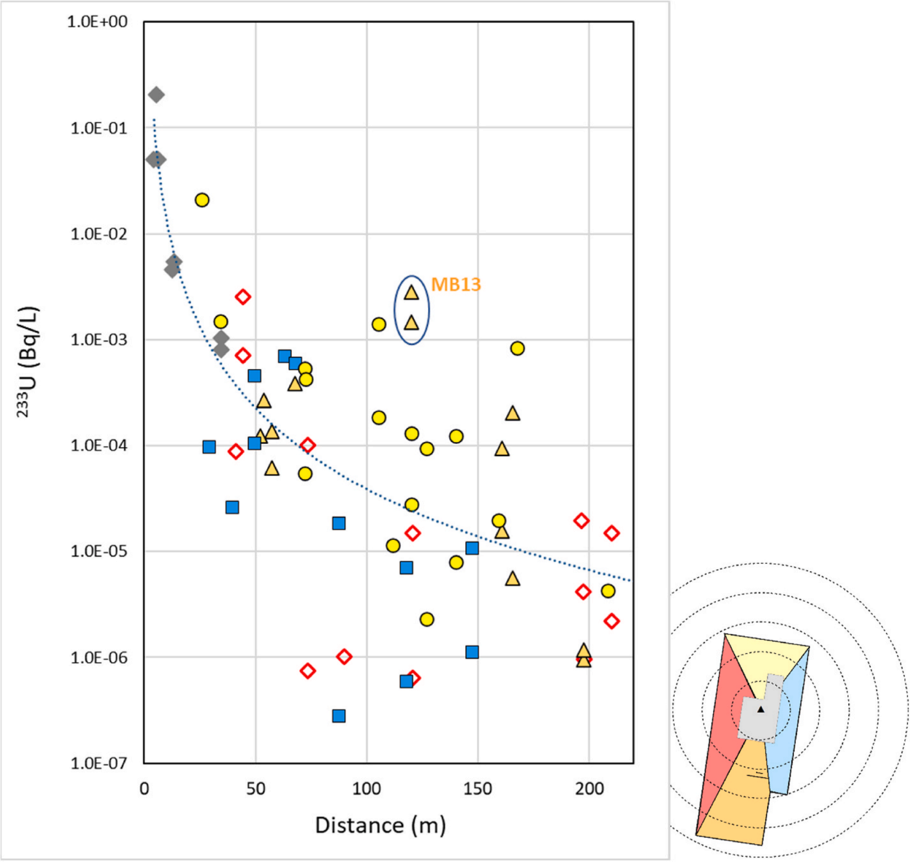

Fig. 4. Activity of ^{233}U in groundwater as a function of radial distance from an origin point (near MB16) in the centre of the trenched area. The grey symbols (diamonds) indicate samples from within, or immediately adjacent to, the trenched area. The western sector (upgradient) is shown by open red diamonds. The colour coding of the other samples is based on direction, as shown in the inset diagram of the site (yellow circles-north, orange triangles-south, blue squares-east). The curve in the main figure is provided for visual guidance. Measurements of ^{233}U in samples from borehole MB13 are indicated. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

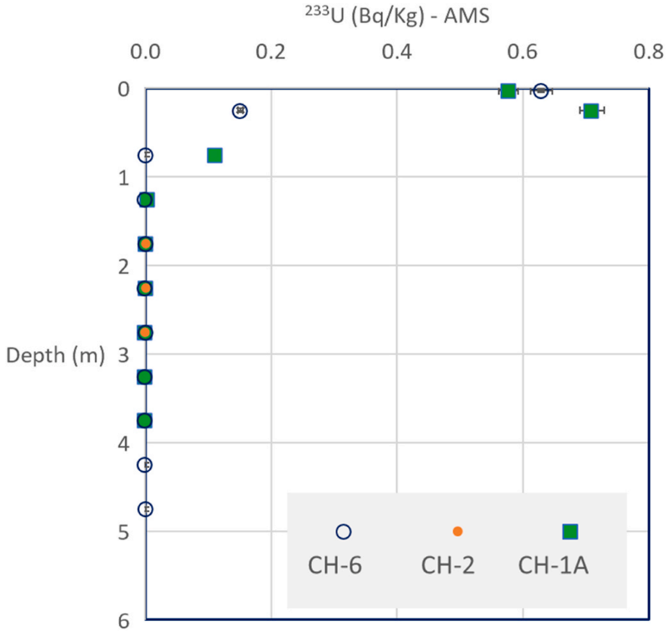

Fig. 5. Depth profiles of ^{233}U in soil samples from the edge of the trenched area (measured by AMS).

of surface or shallow subsurface movement of ^{233}U beyond the trenched area in this direction. However, it cannot be ruled out that the surface soils were contaminated either during waste disposal operations or subsequent activities at the site which may have resulted in dispersion of contamination from the nearby trenched area. There is no evidence in these profiles for deeper (>1 m) subsurface migration of ^{233}U.

4.2. *Surface soils*

It has not been possible within this research project to directly measure ^{233}U in a large number of surface soil samples (other than the soil profile samples discussed above) using AMS. Nevertheless, the spatial data for near-surface soil samples obtained for U isotopes (measured by α-spectrometry) enable us to investigate the possibility that ^{233}U contamination may be present in surface soils across the trenched area.

The distribution map of $^{233+234}$U in surface soils identifies the locations of anomalies in the activities of these U isotopes at the site (Fig. 6a). Additionally, the ratio of $^{233+234}$U/^{238}U activities (Fig. 6b) helps to interpret the data. In Fig. 6b, samples with $^{233+234}$U/^{238}U > 2.0 are depicted in yellow and red. Based on the analogous groundwater data (Fig. 2), it would be expected that these samples may contain elevated ^{233}U. It should be noted that average measured soil activities of ^{238}U and ^{234}U in surface soil samples at the site are 10.5 (±3.5) Bq/kg) and 17.7 (±7.8) Bq/Kg respectively, with the average $^{233+234}$U/^{238}U activity ratio being ~1.7. Therefore, the symbols shown in yellow, orange, and red on both maps indicate data values exceeding these average values, i.e. possible presence of ^{233}U. Comparing both maps,

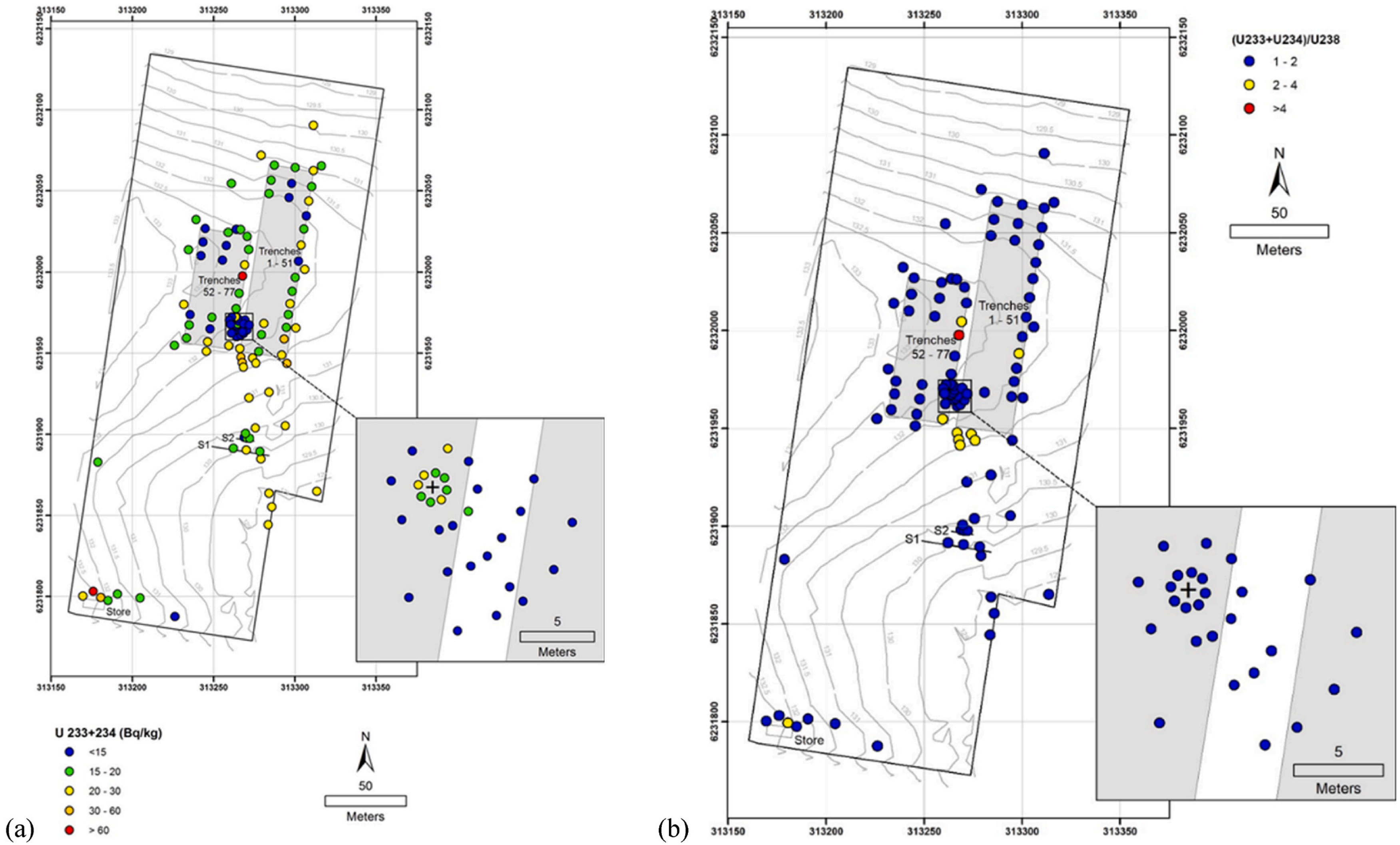

Fig. 6. Alpha spectrometry data for (a) $^{233+234}$U in surface soils (Bq/Kg) and (b) The ratio of $^{233+234}$U/^{238}U activity. The inset squares are expansions of the most Pu-contaminated area of the surface soils, with the Pu derived from an assumed Pu source in the southern part of the western set of trenches (indicated by the + symbol).

significantly elevated values of both the $^{233+234}U/^{238}U$ ratio and the combined $^{233+234}U$ activity were found in a similar location to the anomalous ^{233}U concentrations measured in groundwaters from borehole MB16 in the central part of the trenched area (points shown in red on both maps in Fig. 6).

The inset diagrams in Fig. 6a and b expand an area of the site contaminated with Pu (by the bath-tub effect) from Pu sources buried in this part of the site. Although some samples in this area show elevated total $^{233+234}U$ activity (Fig. 6a, inset), there are no significant increases of the $^{233+234}U/^{238}U$ ratio (Fig. 6b, inset).

Small anomalies in the $^{233+234}U/^{238}U$ ratio are seen in a cluster of samples at the southern end of the trenched area (yellow dots in Fig. 6b). We previously found elevated values of $^{239+240}Pu$ and ^{241}Am in the same area just off the trenches (Payne et al., 2020) and attributed it to surface (or near-surface) movement of contaminants from the main trenched areas. The AMS measurements of ^{233}U in the core samples (described in the previous section) confirmed the presence of ^{233}U in this part of the site near the main trenched area.

Two soil samples with high $^{233+234}U$ activity were found near the decay store at the south-western corner of the site (see Fig. 1). One of these soil samples (red in Fig. 6a) did not have an associated elevated $^{233+234}U/^{238}U$ ratio, hence it seems likely to have resulted from a leakage of natural uranium (containing both ^{238}U and ^{234}U) from a waste item, possibly one of the sludge drums. The possibility of leakage in this vicinity was also suggested by a second sample, showing a slightly elevated $^{233+234}U$ activity (orange in Fig. 6a) and an anomalous $^{233+234}U/^{238}U$ ratio (yellow in Fig. 6b), suggesting this was an item with a higher ^{233}U content. Samples from the same locations near the waste store had previously returned elevated values of both $^{239+240}Pu$ and ^{241}Am (Payne et al., 2020), indicating contamination from leakage of various radionuclides.

The combined data for the groundwaters and surface soil samples suggest that variations of $^{233+234}U/^{238}U$ ratios (Fig. 6b) provide evidence for anomalies in ^{233}U in LFLS surface soils. However, the α-spectrometry results are not conclusive. Therefore, a priority for future work would be to verify the α-spectrometry results by analysing a larger number of surface soils by AMS, focusing on the areas showing elevations in the $^{233+234}U/^{238}U$ ratio.

5. ^{233}U in vegetation

The AMS data for ^{233}U in vegetation samples (Fig. 7) indicates that several samples of both wood and foliage from trees in the vicinity of the LFLS trenches contained measurable amounts of ^{233}U. The samples with higher ^{233}U content also had elevated ratios of $^{233+234}U/^{238}U$ (measured by α-spectrometry), which provides support for using α-spectrometry as a screening method to flag samples of high ^{233}U content. However, some vegetation samples with normal $^{233+234}U/^{238}U$ activity ratios (i.e. 1.0 to 2.0) revealed measurable levels of ^{233}U contamination (Fig. 7). This contamination reflects the uptake of ^{233}U in the trees, noting that all the samples were obtained from trees near the trenches. Similar results have been previously reported for uptake and transpiration of tritium in trees at the LFLS (Twining et al., 2011).

Some of the trees would be expected to have considerable root systems, particularly *syncarpia glomulifera* (commonly known as ‘turpentine’) which can grow to a considerable size exceeding 50 m in height and is thought to have the greatest extent of roots of the sampled trees (Twining et al., 2011). It is likely that the waste trenches are an area of water accumulation and the root system of a tree near to the trenches may be expected to extend through the buried wastes. This is particularly relevant for the *syncarpia* near the south west corner of the trenches (*Sy2*), located close to Trench 55, which contained 0.4 g of ^{233}U disposed in a form described as “aqueous and non-aqueous solutions” (Payne et al., 2022). The location of this trench and this specific tree are indicated in Fig. 1.

While the data on the uptake of ^{233}U in vegetation is limited to a relatively small data set, the results provide confirmation of the presence of ^{233}U in environmental samples from near the LFLS trenches and support the other evidence of movement of ^{233}U through the environment of the trenches.

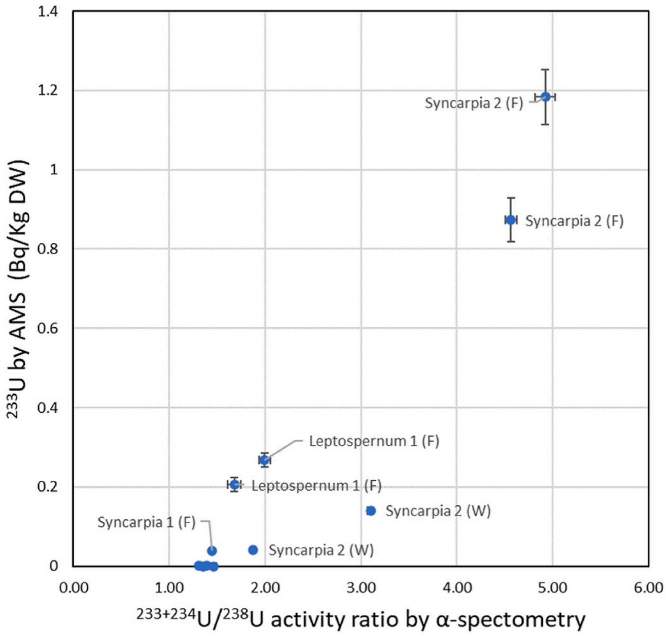

Fig. 7. ^{233}U by AMS and $^{233+234}U/^{238}U$ by α-spectrometry in vegetation samples from the vicinity of the LFLS trenches. The 4 unlabelled data points with negligible ^{233}U are *Eucalyptus*-1 (F) (2 samples), *Acacia*-2 (F), and *Banksia*-1 (F). ‘F’ denotes foliage, ‘W’ is wood.

6. Discussion

The two main anthropogenic isotopes of U which have been reported in environmental samples are ^{233}U and ^{236}U. In general, environmental contamination by ^{233}U is lower than ^{236}U in terms of atom ratios (Chamizo et al., 2022b; Wallner et al., 2022), and the main motivation for using AMS in many studies has been the extremely low detection limits for ^{233}U and the capability to simultaneously measure ^{236}U (Morereau et al., 2022; Tumey et al., 2009). However, in the LFLS study, the main driver for using the AMS technique has been the difficulty of quantifying ^{233}U using α-spectrometry, although it is present in relatively high amounts due to the disposal activities undertaken at the site.

Whilst the levels of ^{233}U in environmental samples (including around power reactors) are usually very low, ^{233}U may be present around nuclear research facilities (Lin et al., 2021). Also, ^{233}U is present in some wastes which may be placed in deep geological disposal (Forsberg and Trabalka, 2001) and ^{233}U has also been reported in tank wastes at the Hanford site (USA) (Christensen et al., 2004). There have apparently been no reports of detection of ^{233}U in environmental samples from Hanford, although anthropogenic ^{236}U as well as the natural U isotopes (Christensen et al., 2004) have been detected. Although there have been some reports of ^{233}U in the environment from nuclear weapons tests (Child and Hotchkis, 2013; Hain et al., 2020); few environmental measurements of ^{233}U from nuclear facilities or waste sites have been reported.

Our results show that ^{233}U is present in environmental samples (soils, groundwater and vegetation) from the vicinity of the LFLS trenches, where ^{233}U is known to have been disposed as part of a short-lived nuclear power program involving ^{233}U fuelled reactors (Hardy, 1999). Numerous other radionuclides were disposed in the trenches and have been detected in environmental samples from the site, including tritium (Hughes et al., 2011), plutonium isotopes (Payne et al., 2013), and several other radionuclides (Payne et al., 2020). The major source of the elevated radionuclide concentrations in the environment near the trenches at this site is the wastes emplaced in the disposal trenches. In the case of ^{233}U, there is evidence that the disposed forms of ^{233}U included potentially mobile powders and liquids, with co-disposal of potential chemical complexants (Payne et al., 2022). Thus, the elevated levels of ^{233}U in environmental samples from LFLS are consistent with the disposal records.

The results for LFLS, where ^{233}U was readily detected by AMS in numerous environmental samples, are considerably different to those obtained for remote environments that are contaminated with trace amounts of U isotopes from nuclear weapons fallout (or from nuclear facilities), where the ^{233}U/^{238}U atom ratio measured in several recent studies ranges from approximately 10^{-12} to 10^{-7} (Chamizo et al., 2022b). The most contaminated groundwater sample for borehole MB16 at LFLS (Fig. 2) corresponds to a ^{233}U/^{238}U atom ratio of $\sim 1.8 \times 10^{-2}$ (approximately 5 orders of magnitude higher than the pristine environments), although the majority of LFLS samples exhibit a ^{233}U/^{238}U atom ratio in the range below 1×10^{-4}.

The reported amount of ^{233}U disposed at LFLS is considered to be an underestimate because the only documented sources of ^{233}U included in the inventory were a small number (~6) of highly contaminated items (Payne et al., 2022). It is probable that additional amounts of ^{233}U were present in other wastes disposed at the LFLS, including a proportion of the approximately 50,000 waste items which were unreported (or incompletely described) in the records. Because of the uncertainty in the reported ^{233}U disposal inventory, a conservatively higher amount of ^{233}U (27.4 GBq) was used in dose modelling related to the current LFLS study (Johansen et al., 2020) (rather than the 1.9 GBq derived from the limited available records). Even with this more conservative approach, the dose from ^{233}U to postulated off-site receptor groups was many orders of magnitude below background. The results indicate that the contribution of ^{233}U to the dose associated with offsite transport from the wastes is not generally significant. However, for specific scenarios where future receptors penetrate into the waste trenches (e.g. a future farmer or resident on the site, burrowing animals, plant roots, and earthworms), the dose from ^{233}U could be comparable to or higher than other U isotopes (Johansen et al., 2020), which confirmed earlier modelling results (Johansen et al., 2012). The implications of this are yet to be tested mainly due to the need for further work on ^{233}U in biota dosimetry (Brown et al., 2016).

7. Conclusions

The results of this study, which were obtained using sensitive AMS measurements, demonstrate that ^{233}U is present in a range of environmental samples from the LFLS, including soils, water and vegetation. The elevated ^{233}U levels in many samples reflect the proximity of trenches in which significant ^{233}U disposals occurred and confirm our extensive evaluation of the disposal records. The findings at LFLS provide important insights into the amounts and environmental behaviour of ^{233}U, for which few previous environmental studies have been reported, particularly in more highly contaminated environments, such as the immediate vicinity of nuclear facilities or legacy sites.

In this study, the combination of historical information from disposal records, together with measurements by α-spectrometry and AMS, has been extremely valuable to fully understand the presence and distribution of ^{233}U at a legacy nuclear waste site. Although the measurement of environmental samples by AMS is a complex and time-consuming exercise, the ^{233}U results by AMS are the most definitive datasets for this radionuclide and provide unique information on the presence and behaviour of ^{233}U at this legacy site.

CRediT authorship contribution statement

Timothy E. Payne: Writing – review & editing, Writing – original draft, Supervision, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Jennifer J. Harrison:** Writing – review & editing, Validation, Methodology, Investigation. **David P. Child:** Validation, Methodology, Investigation. **Stuart Hankin:** Writing – review & editing, Methodology. **Michael A.C. Hotchkis:** Writing – review & editing, Validation, Methodology, Investigation. **Catherine E. Hughes:** Writing – review & editing, Validation, Methodology, Investigation. **Mathew P. Johansen:** Writing – review & editing, Validation, Methodology, Investigation. **Sangeeth Thiruvoth:** Writing – review & editing, Validation, Methodology, Investigation. **Kerry L. Wilsher:** Methodology, Investigation.

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.

Data availability

Data will be made available on request.

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References

AAEC, 1957. Australian Atomic Energy Commission. Annual report. 27–28.

AAEC, 1985. The Little Forest Burial Ground - an information paper. In: Australian Atomic Energy Commission. AAEC/DR19.

Bonhote, P.A., 1964. Possible Concentration and Disposal Processes for Low Level Treatment Plant Sludge Australian Atomic Energy Commission, vol. 253. AAEC/TM.

Brown, J.E., Alfonso, B., Avila, R., Beresford, N.A., Copplestone, D., Hosseini, A., 2016. A new version of the ERICA tool to facilitate impact assessments of radioactivity on wild plants and animals. J. Environ. Radioact. 153, 141–148.

Cendón, D.I., Hughes, C.E., Harrison, J.J., Hankin, S.I., Johansen, M.P., Payne, T.E., Wong, H., Rowling, B., Vine, M., Wilsher, K., Guinea, A., Thiruvoth, S., 2015. Identification of sources and processes in a low-level radioactive waste site adjacent to landfills: groundwater hydrogeochemistry and isotopes. Aust. J. Earth Sci. 62, 123–141.

Cendón, D.I., Rowling, B., Hughes, C.E., Payne, T.E., Hankin, S.I., Harrison, J.J., Peterson, M.A., Stopic, A., Wong, H., Gadd, P., 2022. Rare earth elements and yttrium as tracers of waste/rock-groundwater interactions. Sci. Total Environ. 830, 154706.

Chamizo, E., Christl, M., López-Lora, M., Casacuberta, N., Wefing, A.-M., Kenna, T.C., 2022a. The potential of 233U/236U as a water mass tracer in the Arctic ocean. J. Geophys. Res.: Oceans 127, e2021JC017790.

Chamizo, E., López-Lora, M., Christl, M., 2022b. Performance of the 1 MV accelerator mass spectrometry system at the centro nacional de Aceleradores for the analysis of 233U at environmental levels. Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. Atoms 533, 81–89.

Child, D.P., Hotchkis, M.A.C., 2013. Plutonium and uranium contamination in soils from former nuclear weapon test sites in Australia. Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. Atoms 294, 642–646.

Christensen, J.N., Dresel, P.E., Conrad, M.E., Maher, K., DePaolo, D.J., 2004. Identifying the sources of subsurface contamination at the Hanford site in Washington using high-precision uranium isotopic measurements. Environ. Sci. Technol. 38, 3330–3337.

Forsberg, C.W., Trabalka, J.R., 2001. Disposal of 233U in the Waste Isolation Pilot Plant Waste Management Conference 2001.

Griffiths, H., Payne, T.E., 2018. Management options for the Little Forest Legacy Site, Australia. Radiation Protection in Australasia 35, 3–8.

Hain, K., Steier, P., Froehlich, M.B., Golser, R., Hou, X., Lachner, J., Nomura, T., Qiao, J., Quinto, F., Sakaguchi, A., 2020. 233U/236U signature allows to distinguish environmental emissions of civil nuclear industry from weapons fallout. Nat. Commun. 11, 1275.

Hankin, S., 2012. Little Forest Burial Ground - geology, geophysics and well installation 2009-2010. Australian Nuclear Science and Technology Organisation. ANSTO/E-781.

Hardy, C.J., 1999. Atomic Rise and Fall: the Australian Atomic Energy Commission, 1953-1987. Glen Haven Press.

Harrison, J.J., Payne, T.E., Wilsher, K.L., Thiruvoth, S., Child, D.P., Johansen, M.P., Hotchkis, M.A.C., 2016. Measurement of 233U/234U ratios in contaminated groundwater using alpha spectrometry. J. Environ. Radioact. 151, 537–541.

Harrison, J.J., Zawadzki, A., Chisari, R., Wong, H.K.Y., 2011. Separation and measurement of thorium, plutonium, americium, uranium and strontium in environmental matrices. J. Environ. Radioact. 102, 896–900.

Hoffmann, E., Loosz, T., Ferris, J.M., 2008. Environmental and Effluent Monitoring at ANSTO Sites, 2006-2007. Australian Nuclear Science and Technology Organisation. ANSTO/E-762.

Hotchkis, M.A.C., Child, D., Fink, D., Jacobsen, G.E., Lee, P.J., Mino, N., Smith, A.M., Tuniz, C., 2000. Measurement of U-236 in environmental media. Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. Atoms 172, 659–665.

Hotchkis, M.A.C., Child, D.P., Froehlich, M.B., Wallner, A., Wilcken, K., Williams, M., 2019. Actinides AMS on the VEGA accelerator. Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. Atoms 438, 70–76.

Hotchkis, M.A.C., Child, D.P., Zorko, B., 2010. Actinides AMS for nuclear safeguards and related applications. Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. Atoms 268, 1257–1260.

Hughes, C.E., Cendón, D.I., Harrison, J.J., Hankin, S.I., Johansen, M.P., Payne, T.E., Vine, M., Collins, R.N., Hoffmann, E.L., Loosz, T., 2011. Movement of a tritium plume in shallow groundwater at a legacy low-level radioactive waste disposal site in eastern Australia. J. Environ. Radioact. 102, 943–952.

Johansen, M.P., Barnett, C.L., Beresford, N.A., Brown, J.E., Cerne, M., Howard, B.J., Kamboj, S., Keum, D.K., Smodis, B., Twining, J.R., Vandenhove, H., Batlle, J.V.I., Wood, M.D., Yu, C., 2012. Assessing doses to terrestrial wildlife at a radioactive waste disposal site: inter-comparison of modelling approaches. Sci. Total Environ. 427, 238–246.

Johansen, M.P., Payne, T.E., Comarmond, M.J., Harrison, J.J., Blackley, R., Kabir, A., 2020. Little Forest Legacy Site-Technical Report: Dose Rate Estimates to Humans and Wildlife for a Range of Potential Future Scenarios (ANSTO E-787). Australian Nuclear Science and Technology Organisation.

Leterme, B., Vanclooster, M., Rounsevell, M.D.A., Bogaert, P., 2006. Discriminating between point and non-point sources of atrazine contamination of a sandy aquifer. Sci. Total Environ. 362, 124–142.

Lin, M., Qiao, J., Hou, X., Golser, R., Hain, K., Steier, P., 2021. On the quality control for the determination of ultratrace-level 236U and 233U in environmental samples by accelerator mass spectrometry. Anal. Chem. 93, 3362–3369.

Morereau, A., Jaegler, H., Hain, K., Steier, P., Golser, R., Beaumais, A., Lepage, H., Eyrolle, F., Grosbois, C., Cazala, C., Gourgiotis, A., 2022. Deciphering sources of U contamination using isotope ratio signatures in the Loire River sediments: exploring the relevance of 233U/236U and stable Pb isotope ratios. Chemosphere 307, 135658.

Payne, T.E., 2012. Background Report on the Little Forest Burial Ground Legacy Waste Site. Australian Nuclear Science and Technology Organisation. ANSTO/E-780.

Payne, T.E., Harrison, J.J., Cendon, D.I., Comarmond, M.J., Hankin, S., Hughes, C.E., Johansen, M.P., Kinsela, A., Mokhber-Shahin, L., Silitonga, A., Thiruvoth, S., Wilsher, K.L., 2020. Radionuclide distributions and migration pathways at a legacy radioactive trench disposal site. J. Environ. Radioact. 211, 10681.

Payne, T.E., Harrison, J.J., Hughes, C.E., Johansen, M.P., Thiruvoth, S., Wilsher, K.L., Cendon, D.I., Hankin, S.I., Rowling, B.R., Zawadzki, A., 2013. Trench 'bathtubbing' and surface plutonium contamination at a legacy radioactive waste site. Environ. Sci. Technol. 47, 13284–13293.

Payne, T.E., Shatwell, J., Comarmond, M.J., 2021. Priority issues and key findings from evaluation of disposal records for a legacy radioactive waste site. J. Radiol. Prot. 41, S24.

Payne, T.E., Shatwell, J., Comarmond, M.J., 2022. Waste disposals at the Little forest site – procedures, records and significant events. In: Australian Nuclear Science and Technology Organisation. ANSTO/E-788.

Phillips, J.F., 1967. Solvent Extraction Purification of Uranium-233 with Di-sec-butyl Phenylphosphonate (Report BNWL- 440), Pacific Northwest Laboratory.

Qiao, J., Hain, K., Steier, P., 2020. First dataset of 236U and 233U around the Greenland coast: a 5-year snapshot (2012–2016). Chemosphere 257, 127185.

Rowling, B., Kinsela, A.S., Comarmond, M.J., Hughes, C.E., Harrison, J.J., Johansen, M. P., Payne, T.E., 2017. Measurement of tributyl phosphate (TBP) in groundwater at a legacy radioactive waste site and its possible role in contaminant mobilisation. J. Environ. Radioact. 178–9, 377–384.

Tumey, S.J., Brown, T.A., Buchholz, B.A., Hamilton, T.F., Hutcheon, I.D., Williams, R.W., 2009. Ultra-sensitive measurements of 233U by accelerator mass spectrometry for national security applications. J. Radioanal. Nucl. Chem. 282, 721–724.

Twining, J.R., Hughes, C.E., Harrison, J.J., Hankin, S., Crawford, J., Johansen, M., Dyer, L., 2011. Biotic, temporal and spatial variability of tritium concentrations in transpirate samples collected in the vicinity of a near-surface low-level nuclear waste disposal site and nearby research reactor. J. Environ. Radioact. 102, 551–558.

Wallner, G., Uguz, H., Kern, M., Jirsa, F., Hain, K., 2022. Retrospective determination of fallout radionuclides and 236U/238U, 233U/236U and 240Pu/239Pu atom ratios on air filters from Vienna and Salzburg, Austria. J. Environ. Radioact. 255, 107030.

Wilsher, K., 2012. Radionuclide Bioaccumulation Patterns in Vegetation at a Legacy Low Level Waste Site, Faculty of Science. Charles Sturt University, p. 88.