

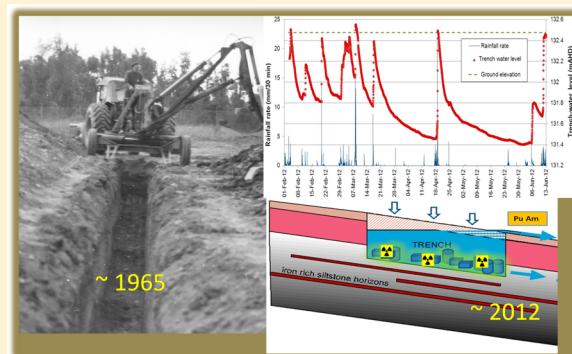
Trench 'Bathtubbing' and Surface Plutonium Contamination at a Legacy Radioactive Waste Site

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Supporting Information

ABSTRACT: Radioactive waste containing a few grams of plutonium (Pu) was disposed between 1960 and 1968 in trenches at the Little Forest Burial Ground (LFBG), near Sydney, Australia. A water sampling point installed in a former trench has enabled the radionuclide content of trench water and the response of the water level to rainfall to be studied. The trench water contains readily measurable Pu activity (~ 12 Bq/L of $^{239+240}\text{Pu}$ in 0.45 μm -filtered water), and there is an associated contamination of Pu in surface soils. The highest $^{239+240}\text{Pu}$ soil activity was 829 Bq/kg in a shallow sample (0–1 cm depth) near the trench sampling point. Away from the trenches, the elevated concentrations of Pu in surface soils extend for tens of meters down-slope. The broader contamination may be partly attributable to dispersion events in the first decade after disposal, after which a layer of soil was added above the trenched area. Since this time, further Pu contamination has occurred near the trench-sampler within this added layer. The water level in the trench-sampler responds quickly to rainfall and intermittently reaches the surface, hence the Pu dispersion is attributed to saturation and overflow of the trenches during extreme rainfall events, referred to as the 'bathtub' effect.



INTRODUCTION

In the years following the Second World War, research into nuclear power and related activities occurred in many countries worldwide. During this period, there was no international consensus on disposal of radioactive waste, and shallow burial in trenches was a commonly used method for disposing of wastes which were deemed to be 'low-level'. While the trench disposal systems were initially believed to be operating satisfactorily, trenches at several disposal locations in the USA were showing evidence of radionuclide movement during the 1960s. There have subsequently been detailed investigations at various legacy sites, such as Maxey Flats, Rocky Flats, Oak Ridge, and other locations in the United States.^{1–4} One of the major concerns has been the mobility of long-lived transuranic actinides such as plutonium (Pu),⁵ americium (Am), and curium (Cm). Significant quantities of Pu were disposed at many near-surface disposal sites, for example, an estimated 80 kg at Maxey Flats,¹ as well as 21 kg at the Beatty facility and 204 kg at a site near Richland.⁶ At some legacy sites, particularly in North America, remediation has been undertaken,⁷ often with extensive public involvement and consultation.⁸ While the most contaminated sites (such as Hanford) have received the most attention, there are examples of USA sites where contamination involving relatively small quantities of Pu (such as an estimated 86 g of Pu at the Rocky Flats '903 Pad') have nevertheless received costly remediation.⁸ As well as remediation of former waste sites, disposal practices at

operational sites have been modified with a greater emphasis on engineered containment, such as at Drigg in the United Kingdom.⁹

Mechanisms of radionuclide mobility and potential remediation actions are central issues in studies of legacy disposal sites. Subsurface pathways are often important, in which radionuclides are released to groundwater and may emerge to the ground surface at some distance from the waste facility.^{3,10} During transit along such a pathway, there is potential for the migration of radionuclides to be mitigated by various retention processes, such as adsorption on mineral surfaces and soil particles. However, at some former trench sites, a phenomena known as the 'bathtub effect' (or 'bathtubbing') has been implicated in the direct release of radionuclides from trenches. This has been described² as a process in which the waste material degraded producing voids within a disposal trench, followed by subsidence of the overlying soil and the entry of surface water into the trench. In cases where the soil surrounding the trenches was sufficiently impermeable, the trenches filled with water. Any overflow of water from this 'bathtub' would have the potential to distribute radionuclides derived from the wastes directly across the surrounding ground

Received: July 23, 2013

Revised: October 7, 2013

Accepted: November 1, 2013

Published: November 20, 2013

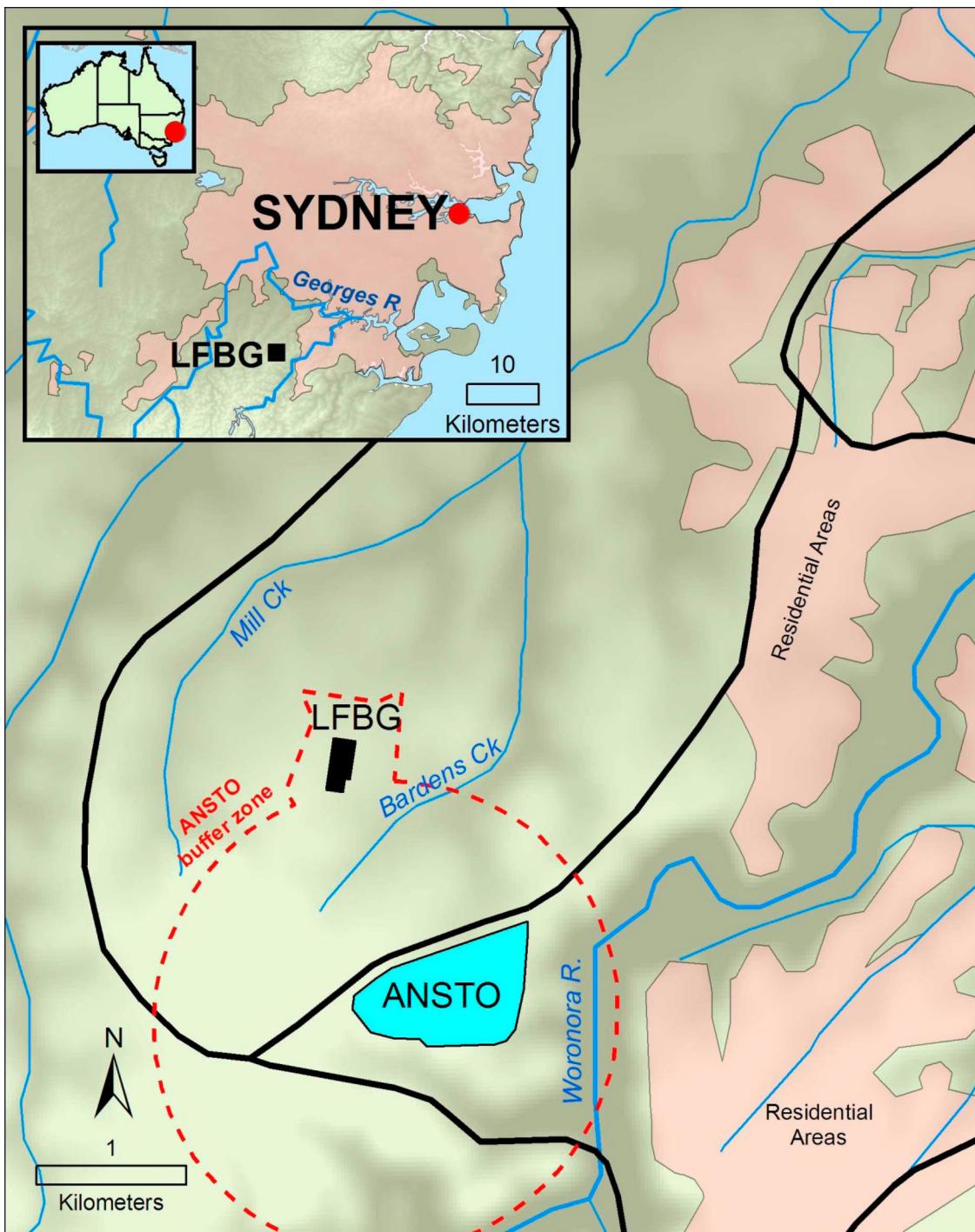


Figure 1. General location of the Little Forest Burial Ground (LFBG). The LFBG occupies an extension of the buffer zone, within which land use is restricted, based on a 1.6 km circle around the former HIFAR reactor.

surface, as appears to have taken place at the Maxey Flats site.¹ While it is considered important to avoid possible ‘bathtubbing’ when designing near-surface waste disposal facilities,¹¹ the mechanism of this process has not been described in detail.

Although radioactive waste sites are not limited to the ~30 countries presently utilizing nuclear power, there have been few reports in the open literature on the status of legacy sites in countries outside North America and western Europe. The present paper concerns the Little Forest Burial Ground (LFBG), which is located near the southern periphery of the city of Sydney in eastern Australia. From 1960 to 1968

radioactive waste, including several grams of Pu, was disposed in a series of trenches at this site by the former Australian Atomic Energy Commission (AAEC). Development of Sydney suburbs in the region near the LFBG has occurred in recent decades, and plans are being considered that may place residences and recreational facilities closer to the LFBG. Therefore it is important to gain a better understanding of the present status, future evolution, and possible management options for the LFBG site.¹²

Site Description and History. The LFBG is situated at 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 from 1958 to 2007 (Figure 1). The trenches were excavated in the surface soil layers above a lens of shale. The vadose zone extends from the surface to a depth of approximately 7–10 m, where the shale layer forms a localized perched water body due to the poor infiltration through the shale. Cores of soils extracted from the vicinity of the trenches during the present project have typically been unsaturated, with variable levels of soil moisture in perched horizons that appear to be discontinuous across the site.¹³ The parent shale is expected to act as a partial barrier to downward movement of groundwater into the aquifer in the underlying Hawkesbury Sandstone. Seeps related to the shale–sandstone interface emerge intermittently at the edge of the shale outcrops around the site.

The waste disposed in the trenches included contaminated items from the operation of the Lucas Heights research facility, waste drums, chemicals, disused equipment, laboratory trash, and beryllium wastes. The LFBG trenches were filled sequentially from 1960 until the cessation of disposal operations in 1968 (see Figure S1 in the Supporting Information). The beryllium (Be) resulted from research into nuclear power systems, and the significant amount of Be disposed (over 1 tonne as both Be and BeO) has been recognized as one of the problematic aspects of the site.¹⁴ Similar to many other disposal sites of this period, the disposal records are incomplete.¹⁵ However, the total activity of radioactive waste was relatively small, for example the Pu disposed was reported as approximately 7 g, which is considerably less than the amounts disposed at the previously mentioned USA sites.

There have been significant changes in land use around the LFBG since the cessation of radioactive waste disposal operations, with major industrial waste and municipal landfill facilities operated in the vicinity. The LFBG site has been fenced and secured, and the AAEC and its successor organization, the Australian Nuclear Science and Technology Organisation (ANSTO), have undertaken ongoing maintenance, surveillance, and monitoring.¹⁶

In the decade immediately following the closure of the LFBG, substantial transfer of various radionuclides from the buried waste to the overlying soil was observed.¹⁴ The evidence for actinide movement was anomalous gross alpha activities up to 22,400 Bq/kg in surface soils above the trenches (recorded in 1974). The gross alpha activity measures all alpha-emitters (both natural and artificial), and for uncontaminated soils of this area was expected to be in the range of 500 to 1000 Bq/kg¹⁴ due to natural uranium and thorium and their decay products. Analyses undertaken at the time by chemical separation followed by alpha spectrometry showed that the elevated gross alpha level was due¹⁴ to the transuranic actinides ²³⁹⁺²⁴⁰Pu and ²⁴¹Am (the α -peaks of ²³⁹Pu and ²⁴⁰Pu overlap and are reported as ²³⁹⁺²⁴⁰Pu). Following these incidents, a further soil layer was added in 1979, and surface levels of gross alpha radioactivity were ‘reduced to normal background levels’.¹⁷

Since this time, 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 LFBG site.^{12,16} A sizable data set has developed for the tritium plume in groundwater, with the data synthesized in a recent paper.¹⁸ Monitoring of γ radiation within the trench area, at a height one meter above ground-

level, consistently indicates no significant difference from background levels. Since the mid 1980s air sampling has been undertaken to detect possible airborne ²³⁹⁺²⁴⁰Pu and Be, which might result from wind dispersion of contaminated surface soil particles. Neither Pu nor Be has been detected throughout this period, except for a possible trace of Pu reported in a composite large volume air sample in 1985.¹⁷

MATERIALS AND METHODS

Field Sampling, Installation of Trench-Sampler, and Analysis. Groundwater samples were obtained from the existing sampling network comprising numerous water sampling wells previously used for environmental monitoring,¹⁶ as well as from new sample wells installed during the present project.¹³ The locations of the sampling wells mentioned in this paper are shown in the Supporting Information (Figure S2).

An additional sampling point, known as the ‘trench-sampler’, was installed when a collapse occurred within the waste disposal area in August 2011, causing a cavity extending from the surface to a depth more than 1.5 m into a former trench. Such collapses have occurred intermittently since the disposal operations and are likely due to the corrosion and collapse of buried objects (such as disposed glove-boxes or waste drums¹²). The 2011 collapse occurred after a period of intense rainfall, and water was visible close to the ground level in the resulting cavity. Following a survey of the area for external radiation dose, a PVC tube-based water sampler was assembled and then carefully lowered into place to avoid damage to any buried objects, and the cavity was backfilled following standard practices for management of the LFBG site. The design of this sampling point is given in Figure S3. This ‘trench-sampler’ was capped to prevent direct entry of rainfall, and an automated logger was installed to provide regular water-level data from the trench.

Groundwater and trench water samples were obtained using plastic tubing and a peristaltic pump. Field parameters were measured using a portable multiprobe system (YSI 556 MPS) with pH/ORP, conductivity/temperature, and DO sensors fitted to a flow cell. The sampling wells were pumped until field parameters were constant or were alternatively pumped until three well volumes were removed (or until dry) then allowed to recharge prior to sampling. The trench-sampler had an ample supply of water, and the trench water level was unaffected by pumping. All wastewater 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 μ m pore-size). Samples were collected and acidified to pH <2 with nitric acid for radionuclide measurement by gamma and alpha spectrometry after standard radiochemical separation procedures.¹⁹ For groundwater samples, 10 L of water was collected for radiochemical analysis, while for trench water samples, 500 mL was sufficient. An unacidified 100 mL sample was also collected for tritium measurement by liquid scintillation using purification and counting procedures described elsewhere.²⁰

Surface soils were generally collected with a push-tube sampler, which obtains a sample of approximately 500 g from the top 10 cm of soil. The soil sampling pattern consisted of points in circles at a radius of 1 and 3 m from the trench-sampler and then samples taken at 6 and 10 m distance in the downhill direction from the trench sampler. Further samples were obtained along two transects away from the trenched area

Table 1. Activity Concentrations of $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am , and Tritium in Trench Water and Nearby Groundwaters^a

	^{241}Am (Bq/L)	$^{239+240}\text{Pu}$ (Bq/L)	^{238}Pu (Bq/L)	^3H (Bq/L)
September 2011 Trench Water				
unfiltered	10.1 ± 0.3	18.5 ± 0.7	0.40 ± 0.03	—
filtered (0.45 μm)	7.3 ± 0.2	12.6 ± 0.5	0.27 ± 0.02	309 ± 6
December 2011 Trench Water				
unfiltered	19.9 ± 0.9	31.0 ± 1.3	0.59 ± 0.04	—
filtered (0.45 μm)	7.4 ± 0.4	11.1 ± 0.2	0.22 ± 0.01	347 ± 6
August 2011 Groundwater from near-Trench Wells (0.45 μm filtered)				
Well W9	0.0023 ± 0.0002	0.0021 ± 0.0002	<0.0002	5315 ± 18
Well W10	0.0038 ± 0.0002	0.0016 ± 0.0001	0.0001 ± 0.0001	8945 ± 23
Well CH30	0.0160 ± 0.0005	0.0215 ± 0.0006	0.0005 ± 0.0001	4188 ± 16

^a ^{238}U , ^{235}U , and ^{234}U in trench waters were <0.05 Bq/L. ^{232}Th and ^{230}Th in trench waters were <0.04 Bq/L. ^{137}Cs and ^{60}Co in trench waters were <0.1 Bq/L. Blank activities were <0.01 Bq/L and <0.0005 Bq/L for alpha-emitters in trench- and groundwater, respectively. The locations of W9, W10, and CH30 are given in Supporting Information, Figure S2.

down the hill-slope toward an ephemeral creek, which receives surface runoff from the site. Numerous samples were also obtained from around the perimeter of the trenched area (full details in Table S1). Several additional shallow surface samples (0–1 cm) were obtained close to the trench-sampler. The soil samples were oven-dried at 60 °C, sieved through a 2 mm mesh, and ground in a ring mill using a carbon steel grinding head. Subsamples of the soils were leached with aqua-regia prior to radiochemical separation and alpha source preparation.¹⁹

Samples from outside the fenced area were independently obtained and analyzed in a separate laboratory to eliminate any possibility of cross-contamination. The first of these samples was obtained in a muddy area close to the fence. The other three samples were sediments from the nearby ephemeral creek which flows into Barden's creek then the Georges River (Figure 1). These samples were predominantly fine-grained, did not require sieving prior to analysis, and were analyzed using similar radiochemical techniques as the samples from within the fenced area.

RESULTS AND DISCUSSION

Actinide Content of Trench Water and Groundwaters.

Two sampling programs were undertaken in September and December 2011 when the water levels were high in the trench-sampler. Preliminary results from gross alpha analysis of the trench water showed unusually high alpha activity. Alpha spectrometry analysis confirmed the presence of significant amounts of $^{239+240}\text{Pu}$ and ^{241}Am in unfiltered trench waters and a lower level of ^{238}Pu (Table 1). While the activities of Pu and Am were reduced by filtration (0.45 μm membrane), the $^{239+240}\text{Pu}$ and ^{241}Am activities in filtered trench water were in the range 7–13 Bq/L. This considerably exceeded Pu and Am activity concentrations measured in filtered LFBG groundwater from wells near the trenches, by several orders of magnitude (Table 1). Activity levels of uranium and thorium were much lower (<0.05 Bq/L).

There are few measurements of similar samples obtained from former trenches in the literature. Much higher $^{239+240}\text{Pu}$ values were reported for water from some trenches at Maxey Flats in the USA, which received liquid wastes that were then solidified, with a resulting pH as low as 1.9 or as high as 12.2. Following these treatments, filtered water (0.05 μm membrane) from those trenches contained $^{239+240}\text{Pu}$ in the range 45–308 Bq/L. In other Maxey Flats trenches with near-neutral pH values (~6–8) the measured $^{239+240}\text{Pu}$ activity in filtered

groundwater was between <0.4 to 13 Bq/L. The $^{239+240}\text{Pu}$ activities in the filtered LFBG trench water, which has a pH of approximately 5.9, are similar to, or slightly higher than, the Pu values for the near-neutral trenches at the Maxey Flats site, where a much greater amount of Pu was disposed. It is possible that our measurements include a significant colloidal contribution between 0.05 and 0.45 μm (due to the different pore-size filters employed). Furthermore, when comparing the measured Pu levels, it should be noted that the Maxey Flats trenches were much larger, hence the Pu disposed there would have been spread over a greater trench volume.

Another plutonium isotope, ^{238}Pu (which has a shorter half-life of 88 years, compared to 24,100 years for ^{239}Pu and 6,540 years for ^{240}Pu), was the highest activity Pu isotope detected in the Maxey Flats trenches (range 36–18500 Bq/L), which was greatly in excess of the $^{239+240}\text{Pu}$ in the same water. This reflected the "significant quantities"¹ of ^{238}Pu which were reportedly disposed at Maxey Flats. The ^{238}Pu concentrations at Maxey Flats were orders of magnitude higher than we report here for the LFBG water (Table 1). This difference in isotopic composition of Pu presumably reflects the different types of waste-generating activities.

Only gross alpha data are available for the Drigg (UK) disposal trenches (rather than individual actinides), with the maximum values for unfiltered water from within the Drigg trenches being around 0.67 Bq/L.²¹ The LFBG trench-sampler results (Table 1), which amount to as much as 50 Bq/L for the sum of ^{241}Am and $^{239+240}\text{Pu}$ in unfiltered trench water, are much higher, even without considering any other alpha-emitters.

The presence of ^{241}Am in the LFBG trench water samples (Table 1) possibly indicates similar mobilization mechanisms to Pu. Americium was also mobile in conjunction with Pu, at Mortandad Canyon (Los Alamos National Laboratory),²² and ^{241}Am was also released at Oak Ridge³, where it was predominantly associated with the transuranic actinide isotope ^{244}Cm .

There has been a major international study of trenches containing contaminated materials from the Chernobyl accident, which has resulted in a series of papers.^{10,23,24} The Chernobyl trenches were located above the usual water table and underlain by sands with relatively high hydraulic conductivity.¹⁰ Therefore, water might not accumulate in the trenches and the collection of in-trench samples may not be possible there. Outside the Chernobyl trenches, the $^{239+240}\text{Pu}$ activities in groundwater samples were below 1 Bq/L.¹⁰

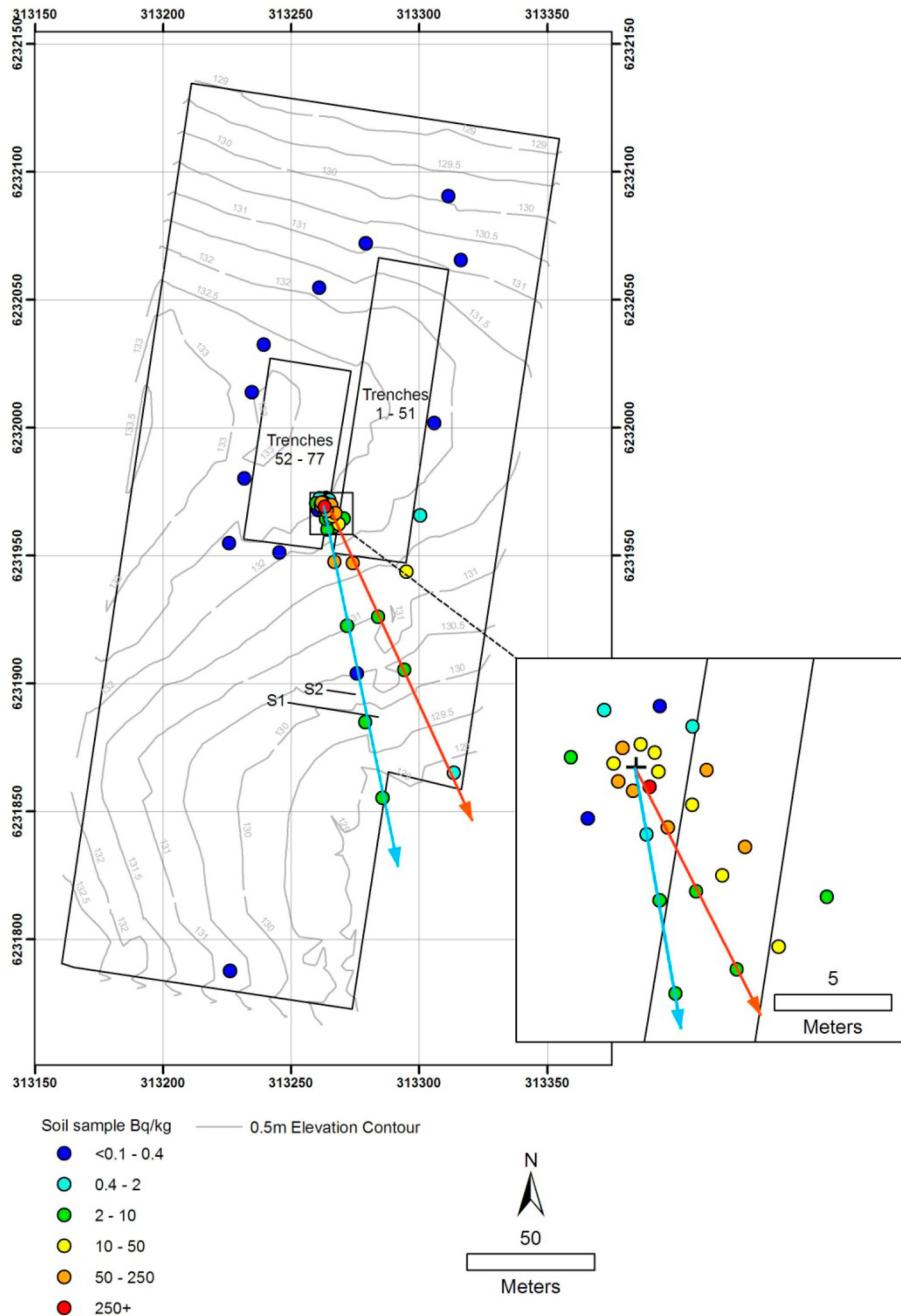


Figure 2. Distribution of surface soil plutonium activities (0–10 cm depth) at the LFBG (samples taken on 15 December 2011). The main trenched areas are in the two rectangles in the center of the figure, comprising an eastern set (trenches 1–51) and a western set (trenches 52–77). Two additional trenches (S1 and S2) were located to the south of the main trenched areas. These trenches are intersected by the light blue transect, which is referred to as the 'MB13 transect' (borehole MB13 lies on this transect). Surface elevation contours indicate the general direction of surface water flow. Full Pu data are given in the Supporting Information (Table S1). The inset to the lower right of the figure expands the area nearest the trench-sampler (indicated by +).

The levels of $^{239+240}\text{Pu}$ in LFBG trench waters are therefore relatively high, based on the limited comparable data in the literature for samples from similar trench environments. However, our samples were obtained directly from a waste trench in a site containing known Pu contamination and probably reflect the presence of a nearby buried source within the trench. Although the LFBG disposal records are not complete, they indicate that the trenches near the trench-sampler likely received a major proportion of the Pu disposed at the site.¹² The trench water actinide data might also reflect unfavorable geochemistry, or the presence of codisposed

complexing contaminants, as was implicated in the mobilization of Pu at Maxey Flats.¹ We are presently undertaking detailed chemical investigations of the trench water and developing methods of separating the oxidation states of Pu and Am, so that their chemical speciation in the trench water can be investigated.

The analyses for the nonactinide contaminants show relatively low levels of gamma-emitting radionuclides in the trench water (Table 1), and no significant amounts of beryllium (<0.01 mg/L). The tritium level is also relatively low, given that the LFBG trenches are a source of tritium contamination and

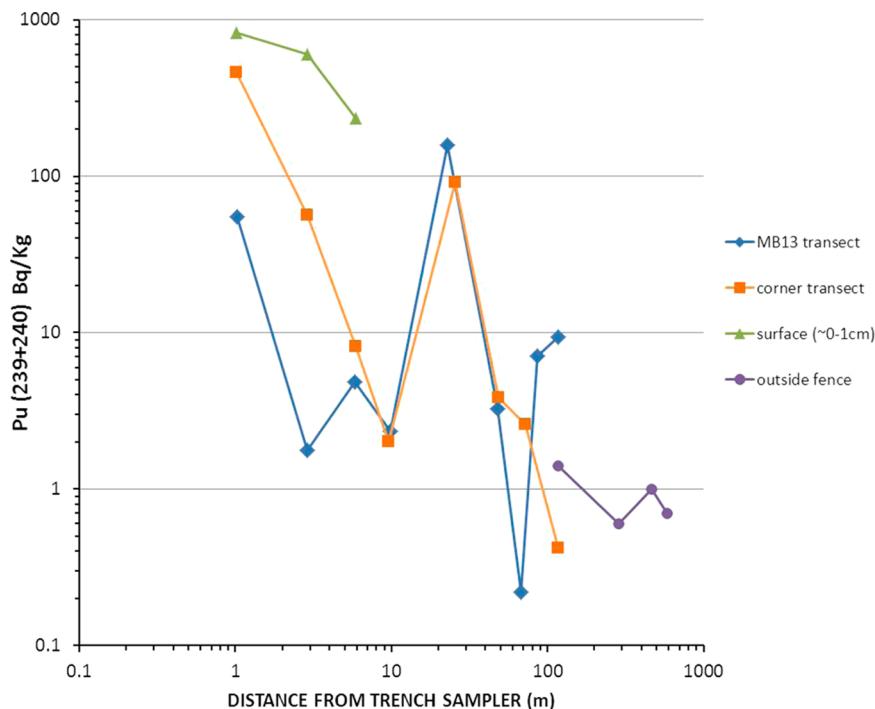


Figure 3. Plutonium activities along linear transects from the trench collapse location. The blue ('MB13') and orange ('corner') transects are the same as in Figure 2. Note the higher concentration in the shallow (0–1 cm) samples, the apparent peaks in soil Pu at the edge of the trenched area (~25 m from the trench sampler), and also the increase where the MB13 transect crosses the two isolated trenches S1 and S2. The fence-line is around 100 m from the trench-sampler at the end of the orange and blue transects. The first sample beyond the fenced area is within 2 m of the final sample on the blue transect. The 3 most distant samples beyond the fenced area (>250 m from the trench-sampler) are sediments from the ephemeral creek.

that groundwater from the immediate vicinity typically shows elevated tritium contents¹⁸ (see Table 1). Tritium is extremely mobile, and it appears that either there was not a significant amount of tritium disposed in this trench, or it has been diluted by rainwater and washed out during the subsequent years.

Plutonium Distributions in Surface Soils. The spatial data on the measured activity of $^{239+240}\text{Pu}$ in surface soils is presented in Figure 2. The density of sampling points was not sufficient for contouring of the measured Pu activities, hence colored symbols are used to represent the measured values. In this diagram, the lowest $^{239+240}\text{Pu}$ activity concentrations (<0.4 Bq/kg) are represented by dark blue markers, and these occur around much of the western and northern perimeter of the trenched area. Although the regional background level of Pu in soils of the Sydney area has not been determined, an average value for $^{239+240}\text{Pu}$ of 0.356 Bq/kg (with a range from 0.055 to 1.003 Bq/kg) was recently reported in 11 surface soils and sediment samples taken from locations in the Australian Capital Territory and New South Wales.²⁵ These locations, within a few hundred kilometers of the LFBG, are assumed to be affected by fallout from nuclear weapons tests to a similar extent. Thus, we conclude that many soil samples from around the perimeter of the LFBG trenched area do not exhibit significantly elevated Pu concentrations compared to soil samples from elsewhere in eastern Australia.

The highest activity of $^{239+240}\text{Pu}$ in 0–10 cm surface soil (represented by a red symbol in Figure 2) was 458 Bq/kg in the immediate vicinity of the trench-sampler, showing clear evidence of contamination from the trenches. This result is substantially elevated relative to surrounding soils but is considerably lower than the value of 22,400 Bq/kg (gross alpha) reported for a soil sample taken in 1974 prior to the

addition of the surface soil layer.¹⁴ Thus, it appears that the added soil layer has been partially effective in restricting the migration of Pu from the trenches. Nevertheless our highest soil value, exceeding background by 3 orders of magnitude, must reflect contamination of the layer which was added in 1979.

The map of the $^{239+240}\text{Pu}$ activity in the surface soils shows surface contamination extending from the vicinity of the trench-sampling point down the slope within the fenced area (Figure 2). The contamination appears to originate from a position close to the trench-sampler, although this is not necessarily the only source of $^{239+240}\text{Pu}$ contamination. On the trenched area the surface contamination is somewhat unevenly distributed (see inset to Figure 2). This may represent meandering flow-paths on the surface or possibly results from water movement in the shallow subsurface, which causes the contamination to come to the surface unevenly in localized parts of the trenched area.

A more detailed representation of the activity of $^{239+240}\text{Pu}$ along the sampling transects is shown in Figure 3. It appears that the Pu measurements on the two transects do not decrease uniformly, instead they fall steeply within 10 m of the trench-sampler and then increase in a clear spike just off the trenched area (this elevated region is visible in Figures 2 and 3). It is not possible to delineate a surface pathway of elevated $^{239+240}\text{Pu}$ from the trench-sampler to these locations on the edge of the trenched area. As noted above, this may be because of meandering flow-paths or due to subsurface flow through the added fill layer on the surface, which is re-emerging at the edge of the trenched area. It is also possible that the contamination beyond the trenched area is a remnant of the contamination

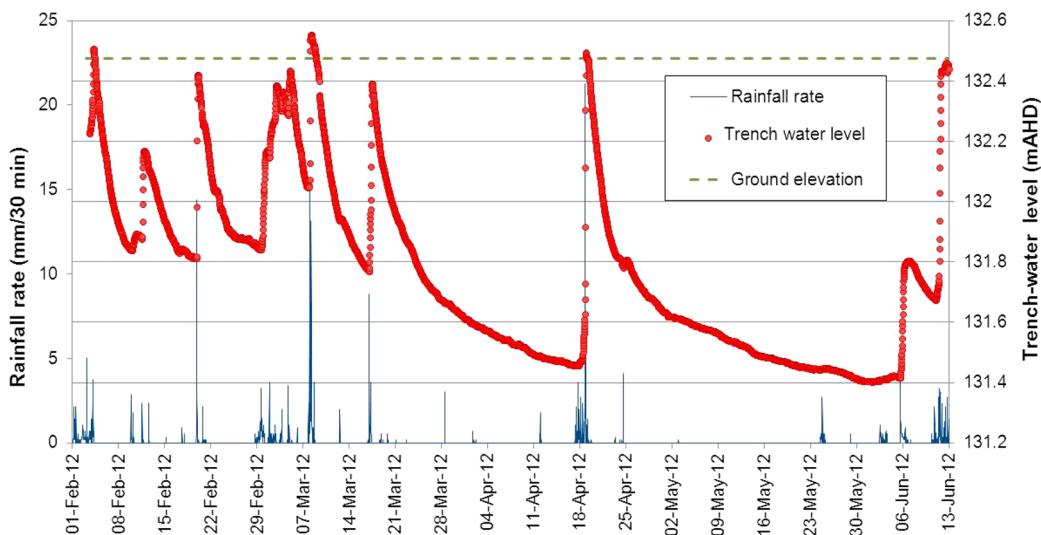


Figure 4. Water level in the trench-sampler from February to June 2012, showing the response of the water levels (with respect to Australian Height Datum) to rainfall. Ground level is indicated by the horizontal dashed line.

detected in the 1970s, although we have not located any previous report of contamination away from the trenches.

Figure 3 also shows the data for the samples from 0 to 1 cm depth taken from the immediate vicinity of the trench-sampler. These shallow samples exhibit higher Pu activity concentrations ($^{239+240}\text{Pu}$ of up to 829 Bq/kg) compared to the standard samples which represent the top 10 cm. The presence of higher Pu in these shallow samples from the top layer of surface soils is a further indication of a surface transport pathway.

Away from the most contaminated area near the trenches, the activity of Pu on the two transects decreases toward the fenceline. However, the Pu concentrations are slightly elevated at the fence, relative to the lowest samples on the perimeters of the trenched area away from the presumed flowpath. The transect past borehole MB13 (shown in blue in Figure 3) exhibits an apparent spike of $^{239+240}\text{Pu}$ around 100 m from the trench-sampler. At this distance, this transect crosses the isolated trenches S1 and S2, and it is possible that these contain another source of plutonium contamination. There is incomplete documentation of the contents of trenches S1 and S2, which were filled at a time when plutonium was being routinely handled in the AAEC research laboratories.¹²

The levels of $^{239+240}\text{Pu}$ in the three sediment samples from the ephemeral creek draining the site are in the range from 0.6–1.0 Bq/kg, which exceeds the lowest values reported in the soils around the trenched area. These sediment $^{239+240}\text{Pu}$ levels are within but toward the upper end of the range (0.055–1.003 Bq/kg) reported for the soil and sediment samples from nearby parts of Australia²⁵ (discussed above) and also exceed (by a factor of over two) the mean of those previously reported values. Compared to the local soils, the creek sediments include greater amounts of fine organic and inorganic materials, which can often concentrate contaminants, due in part to their higher surface area per unit volume.^{5,26} It is therefore possible that the sediments contain traces of Pu contamination from the LFBG wastes, and further isotopic studies (particularly $^{239}\text{Pu}/^{240}\text{Pu}$ ratios) will be needed to determine whether part of their Pu content was derived from the LFBG source as well as from fallout.

It has previously been shown that there is ongoing release of tritium from the LFBG trenches through the subsurface, in

which tritiated water (which is chemically the same as water) is not retarded significantly.¹⁸ Despite the presence of substantially elevated tritium in many samples, groundwater from near the trenched area (excluding the trench water) contains relatively low levels of $^{239+240}\text{Pu}$ (Table 1). Thus, we conclude that, in contrast to the tritium, the actinides are mostly dispersed via a surface route. This Pu migration route is similar to that proposed for Pu movement in the Mortandad Canyon, Los Alamos by Marty et al.,²⁷ who reported that Pu moved via surface pathways twice as fast as groundwater flow and outdistanced the tritium peak in the groundwater. A significant implication of these findings is that tritium, despite its mobility, cannot always be relied on as an advance indicator of the movement of other radionuclides (such as Pu) in environmental pathways at LFBG and other radioactively contaminated sites.

Trench Water Level Response to Rainfall and the ‘Bathtub’ Effect. The installation of a water-level logger in the trench sampler has enabled the collection of a data set for the water level in the trench and its response to rainfall (Figure 4). During the early part of 2012, which was an unusually wet period, the logger recorded several instances of saturation and subsequent overflowing of the trench during a series of frequent and intense rainfall events. This clearly provides a mechanism for surface contamination of radionuclides. Later in 2012, the rainfall patterns became less regular and trench water levels gradually decreased. These observations provide a valuable insight into the behavior of a legacy trench site, particularly in clay-rich soils in this type of climate, and provide evidence of ‘bathtubbing’ within the trench.

Sydney is subjected to periods of prolonged droughts, and in dry periods the trench water level at LFBG may be below the ground surface for long periods. The water level only reaches the ground surface infrequently and briefly during major rainfall events and then recedes during dry periods. The lack of any response of water levels in the trench-sampler to pumping reflects the large volume of available water when the water-level is in the zone accessible by the trench sampler. The trench water level rises to near the surface within hours in response to high intensity rainfall (Figure 4) then decreases relatively rapidly by approximately 0.5 m over 4 days after rain ceases. A

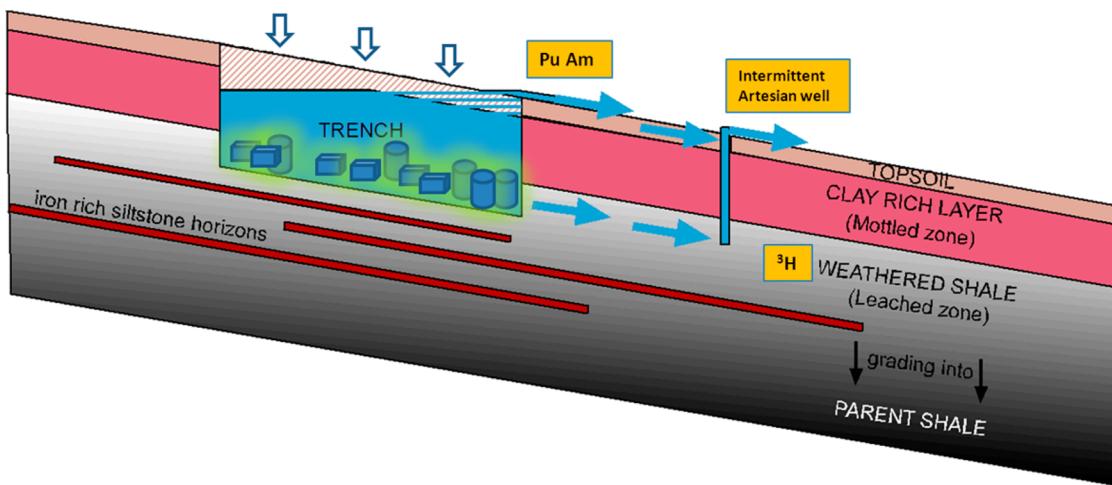


Figure 5. Conceptual model of a trench at the LFBG. Note that the slope of the trench is exaggerated. The main feature described in the present paper is the overflowing at the end of the trenches when the water level is high.

slower decrease is then observed if there is no further rainfall. Thus, it appears that there is a continued release of water into the shallow subsurface when the water level is high, particularly into the unconsolidated layers above the former trenches.

A conceptual model of the trenches (Figure 5) explains a number of the observed features of the site, specifically the surface expression of actinides near the trenches. The trenches are slightly inclined along their long axes, with the eastern ends ~0.4 m below the western end. The thick clay layer is responsible for holding the water in the trench for long periods. The clay soil at LFBG, which has relatively favorable ion-exchange and sorption properties,¹⁴ appears to be inhibiting migration of many radionuclides (other than tritium) through the subsurface pathways. Therefore, the plutonium concentration in groundwater samples from areas close to the trenches is much lower than in trench waters (Table 1).

The low permeability of the clay-rich soils is the major factor causing the 'bathtub' effect at the site, leading to surface release of trench-water, which has elevated Pu and Am levels (Table 1). Unlike the clay layers in the shallow subsurface, the soil above the trench is disturbed and relatively permeable to water. Similarly to a bathtub, the former trench fills with water and overflows when rainfall is sufficient. The overflows at the eastern end of the trenches have led to the dispersion of Pu to nearby surface soils.

Figure 5 also shows the apparent subsurface pathway below the clay layer in which the tritium appears to migrate (and in which the Pu movement is significantly retarded), which explains observations of artesian behavior in some wells (see Figure S4). The diagram also shows the shallow surface layer which was added in 1979 following the earlier contamination events. Due to seepage through this shallow surface layer following major rainfall events, there may be higher actinide concentrations within this layer.

Implications for Management of LFBG and Similar Sites. This paper has shown that the 'bathtub' effect is occurring in trenches at the LFBG legacy waste site, which contain water with elevated levels of both $^{239+240}\text{Pu}$ and ^{241}Am . This has led to a dispersion of plutonium from the buried items into surface soils. At least part of the release of $^{239+240}\text{Pu}$ from the LFBG trenches has occurred since the emplacement of the soil layer which was added in 1979 following earlier surface contamination events. Although we have established a possible

mechanism for off-site surface transport of contamination from LFBG, the levels of $^{239+240}\text{Pu}$ in creek sediments (around 0.6 to 1.0 Bq/kg as $^{239+240}\text{Pu}$) draining the affected area are not above the range of similar samples from nearby parts of Australia derived from fallout²⁵ (0.05 to 1.0 Bq/kg), although, as discussed above, they are at the upper end of this range. Values for Pu in samples from Europe, affected by different fallout patterns (and contributions from the Chernobyl accident) can be somewhat higher, with a survey of soils reporting $^{239+240}\text{Pu}$ between 0.04 and 8.27 Bq/kg in Montenegro²⁸ and surface samples from Kosovo were also around 1.0 Bq/kg.²⁹

The significantly elevated Pu soil activities at LFBG are localized within the fenced area containing the former trenches and are much lower than typical values measured at the Maralinga nuclear test site prior to its remediation.³⁰ A direct comparison of the measured Pu levels at LFBG with the cleanup criteria adopted for the Maralinga site is difficult because the criteria for Maralinga were location-dependent and based on ^{241}Am rather than plutonium.^{31,32} However, measured Pu concentrations in LFBG soils are below the cleanup levels adopted for overseas nuclear legacy sites, even where access is unrestricted. These cleanup levels are typically framed in terms of a median or average plutonium activity concentration for surface soil samples, determined over a small area, rather than peak activities. For example, a cleanup criteria for the former nuclear weapons test-site at Enewetak Atoll, in the Marshall Islands, was reported as 1480 Bq/kg of total transurans (averaged over 0.25 ha) for residential islands and up to 4 times higher for nonresidential islands.³¹ A "soil action level" of 1850 Bq/kg plutonium was established in 2002 at Rocky Flats (site 7), located near the city of Denver, USA.⁸ Similar to the LFBG, this site has been subjected to encroachment by residential suburbs. Prior to cleanup, extensive sampling of Rocky Flats soils, which involved compositing 25 evenly spaced samples, yielded a mean $^{239+240}\text{Pu}$ value of 1924 Bq/kg.⁴ These reported average levels, and the eventual cleanup levels, are above the maximum measured $^{239+240}\text{Pu}$ level at the LFBG of 829 Bq/kg in 0–1 cm surface soil (Table S1) and considerably exceed the median value on the LFBG trenched area within 10 m of the trench-sampler (31.7 Bq/kg). Furthermore, the Pu concentrations at LFBG were generally lower off the trenched area. Therefore, the surface levels of plutonium which have been measured at LFBG would not warrant remediation if similar

standards to Rocky Flats (or Enewetak) were applied, particularly given that the LFBG site is fenced and access is controlled.

However, the levels of Pu in the most contaminated LFBG soil samples exceed the IAEA clearance level (100 Bq/kg) for release of bulk materials, below which regulatory controls are not required.³³ They also exceed the generic USEPA soil screening level (SSL) guidance of ~100 Bq/kg (soil ingestion pathway)³⁴ which represent an indicative threshold for further investigation and is based on conservative exposure assumptions to a hypothetical future occupant of the site. The exceedance of these screening values indicates the need for additional assessment and for continued monitoring and control, together with evaluation of possible interim and long-term remediation options. Given the proximity of a major suburban area, a precautionary approach is justified. Based on the results presented here, any intervention would aim to address the 'bathtub' effect and minimize radionuclide dispersion from the trenches, particularly movement toward surface catchments.

The plutonium disposed at LFBG was derived from research into nuclear power, which has never been used in Australia. Our findings may have implications for similar legacy trench sites elsewhere, even in nations without nuclear power where only relatively small amounts of radioactivity may have been disposed. The trench-sampler described in this paper enabled quantification of actinides in water within a former waste trench, as well as providing the capability to monitor the fluctuating water levels and their response to rainfall. These measurements, together with the delineation of surface plutonium contamination, yielded new insights regarding the phenomena known as the 'bathtub' effect and its consequences in facilitating the spread of contamination from disposal trenches.

ASSOCIATED CONTENT

Supporting Information

Figure S1 – the sequence of filling the trenches at the LFBG from 1960 to 1968, superimposed on an original plan of the site. Figure S2 – groundwater sampling wells at the LFBG. Figure S3 – construction of the trench-sampler installed at the LFBG. Figure S4 - photograph of well OS3 at the LFBG during a time of high water levels. Table S1 – measured ²³⁹⁺²⁴⁰Pu activity concentrations in LFBG surface soils. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This research was conducted within the Nuclear Methods in Earth Systems Project of ANSTO. Numerous members of this project participated in field sampling, data interpretation and sample analysis. The contributions of other ANSTO staff across the organization are gratefully acknowledged, particularly those from the environmental monitoring group, waste operations, health physics, records, and the Institute for Environmental Research.

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