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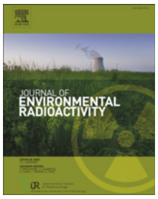

Movement of a tritium plume in shallow groundwater at a legacy low-level radioactive waste disposal site in eastern Australia

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ABSTRACT

Between 1960 and 1968 low-level radioactive waste was buried in a series of shallow trenches near the Lucas Heights facility, south of Sydney, Australia. Groundwater monitoring carried out since the mid 1970s indicates that with the exception of tritium, no radioactivity above typical background levels has been detected outside the immediate vicinity of the trenches. The maximum tritium level detected in groundwater was 390 kBq/L and the median value was 5400 Bq/L, decay corrected to the time of disposal. Since 1968, a plume of tritiated water has migrated from the disposal trenches and extends at least 100 m from the source area. Tritium in rainfall is negligible, however leachate from an adjacent landfill represents a significant additional tritium source. Study data indicate variation in concentration levels and plume distribution in response to wet and dry climatic periods and have been used to determine pathways for tritium migration through the subsurface.

1. Introduction

Tritium (^{3}H) is a common groundwater contaminant at sites associated with a range of human activities such as nuclear power generation, research reactors, nuclear weapons and disposal of radioactive, industrial and urban waste in landfills. The Hi-Flux Australian Reactor "HIFAR" commenced operation in 1958 at the Lucas Heights facility of the Australian Atomic Energy Commission "AAEC" (Fig. 1). The nearby Little Forest Burial Ground (LFBG), located 1.8 km north of HIFAR, was used for the disposal of some of the low-level radioactive waste generated at the Lucas Heights research facility in a series of shallow trenches. Disposal of waste at the LFBG ceased in 1968 and the site has been routinely monitored since that time, with the results published annually in facility reports (see Supplementary Data Table S1). Shallow burial in unlined trenches has been considered to be an appropriate method for the disposal of low-level radioactive waste (IAEA, 1999) subject to siting considerations to prevent unacceptable levels of groundwater contamination or movement of contaminants from the site to the accessible environment. The amount of waste emplaced at LFBG is relatively small, and the monitoring has shown that there are no significant exposures to members of the public from the waste under the current management regime. However, a research study of LFBG has commenced in order to better understand the present status and expected evolution of contaminant migration at the site. This paper uses over 30 years of groundwater tritium data from the Little Forest Burial Ground to assess the potential transport of mobile radionuclides from the site and to develop a conceptual model of the local hydrogeology. This research is intended to provide a scientific basis for decisions concerning possible future management options which may include in situ remediation, exhumation, or maintenance of the present management regime.

Tritium is often considered to be non-sorbing (conservative) during movement through porous media and will be considered so in this paper. This attribute, and the relatively low levels at which it can be reliably measured (IAEA, 2001), make tritium one of the most widely used tracers in groundwater studies of contaminated and natural sites (e.g. Egboka et al., 1983; Goutal et al., 2008; Hackley et al., 1996; Hu et al., 2005, 2008; Kerfoot et al., 2003; Mitchell et al., 2005; Zhang et al., 1995). However, recent studies suggest that the movement of tritium may be attenuated by clay minerals (López-Galindo et al., 2008; Kalinichenko et al., 2008).

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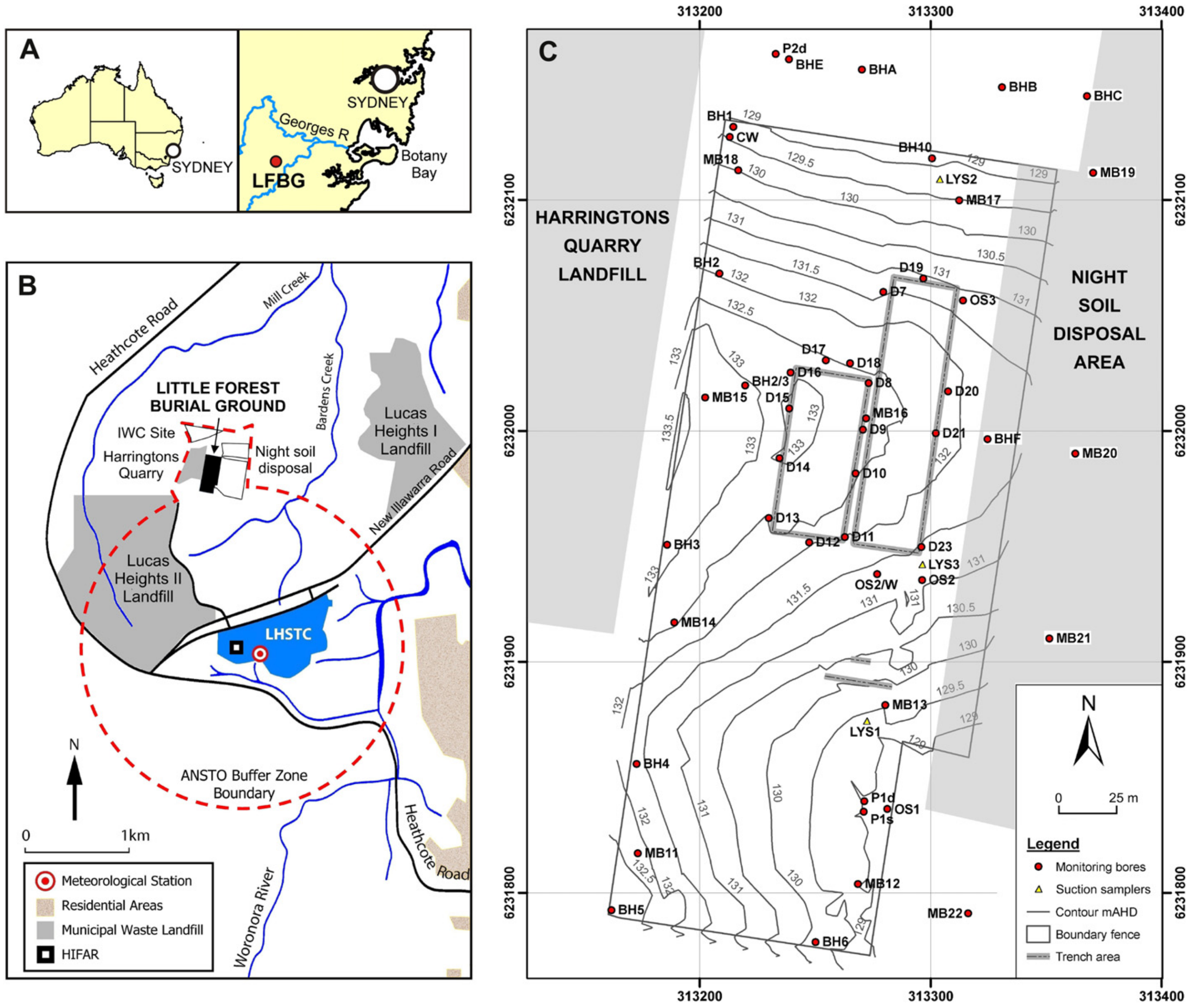

Fig. 1. A. Little Forest Burial Ground location map, Sydney, NSW, Australia Lat 34 2′10″S Long 150 58′ 58″E; B. Surrounding land uses; C. Borehole locations and contour map of LFBG.

2. Study site description

2.1. Physiography and geology

The southern Sydney locality of Lucas Heights is situated on the south-eastern edge of the Georges River catchment, which flows into Botany Bay (Fig. 1A). The LFBG site is a 5.5 ha fenced area within the ANSTO buffer zone around the Lucas Heights Science and Technology Centre, located near the top of the catchment on the Woronora Plateau. The disposal area is situated on a water divide between Mill and Barden Creeks. Surface runoff from LFBG flows both north towards Mill Creek, and south-east towards Barden Creek (Fig. 1B).

The dominant lithologies outcropping on the Woronora Plateau are of Triassic age and consist of the Hawkesbury Sandstone which extends to a depth of approximately 200 m (Sherwin and Holmes, 1986), with conformably overlying shallow remnants of shale and thin sandstone and siltstone lenses (Fig. 2), which has been referred to as the Ashfield Shale (Isaacs and Mears, 1977).

The soils of the site are highly weathered and derived from the shale layer which is generally weathered to a depth of ~6 m, resulting in a profile from surficial red–brown clay soil, through mottled grey and silty kaolinite clay with numerous fine sandy partings. Immediately below the soil and weathered material lies light grey leached silty shale grading to fresh dark carbonaceous parent shale with silty interbedding. This parent shale grades into shale/sandstone laminae above the Hawkesbury Sandstone. From the centre of the trench area extending northwards, siltstone has been observed in drill cuttings, suggesting a discontinuous siltstone layer or lens increasing in thickness at a depth of ~2 m. The soil and shale layer ranges from 7 m to >14 m thick at the burial trenches, thinning to as little as 4 m on the south-eastern periphery (AAEC, 1966). The shale lens extends beyond the LFBG site boundary to the east and west (Knight et al., 1978; Bradd, 2003), and has been actively mined in the immediate vicinity of LFBG.

The Hawkesbury Sandstone consists primarily of laterally discontinuous quartzose sandstone sets ranging from 1.5 m to 3.0 m in thickness, with some siltstone/fine sandstone laminae, siltstone and claystone interbeds. The structure of the sandstone varies from massive to cross-bedded. Inspection of cores obtained in the local area infers relatively high permeability in some of the coarser grained, less cemented sandstone. Weathering occurs

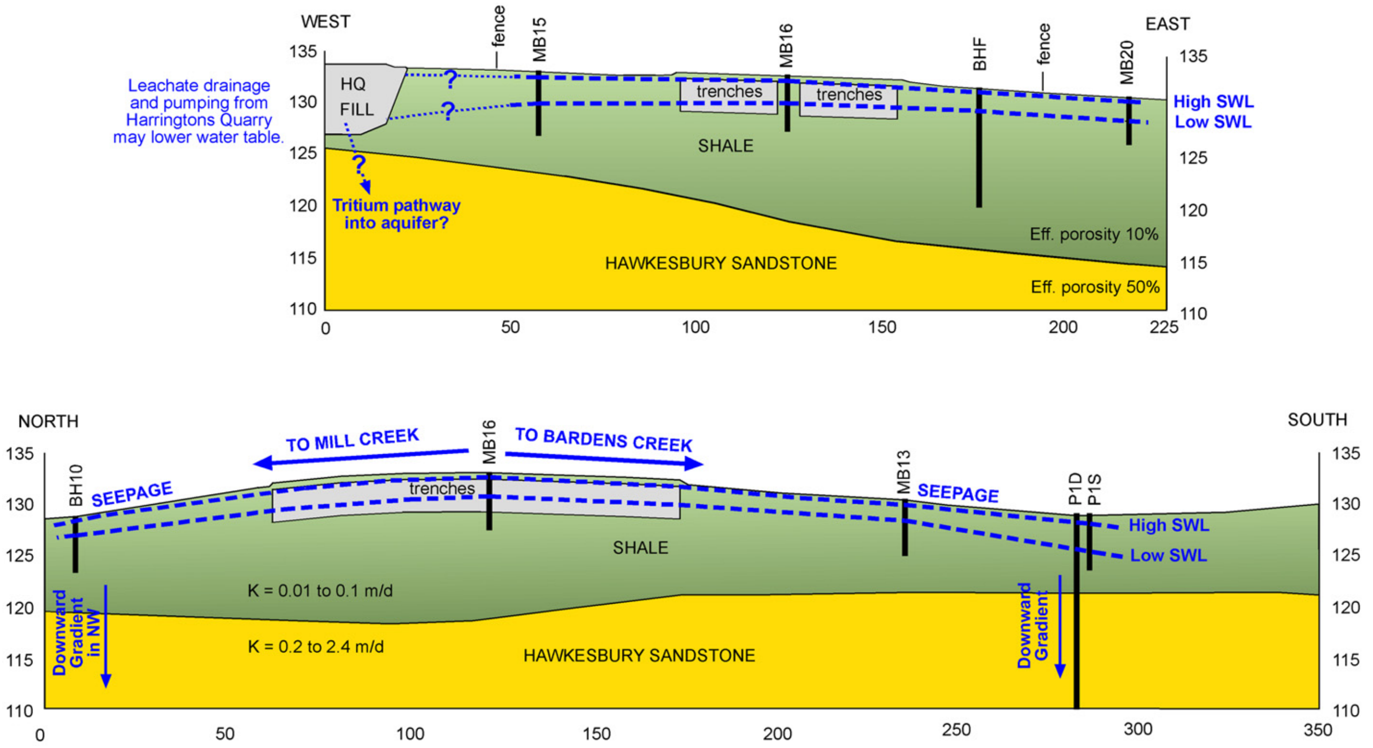

Fig. 2. Conceptual model of Little Forest Burial Ground hydrogeology illustrated using W–E and N–S transects showing the location of LFBG trenches and Harringtons quarry. The range of water table levels in relation to the trenches, surface discharge/seepage zones, hydraulic properties and hypothesised flow pathways are indicated.

predominantly along joint sets increasing secondary porosity and therefore water transmissivity.

2.2. Climate

The LFBG area has a warm temperate climate with maritime influences. There are prolonged periods of high temperature, with the warmest month (February) having average maximum and minimum temperatures of 25.9 °C and 17.4 °C respectively, with daytime maxima over 40 °C being common. The coldest month is July with maximum and minimum temperatures of 15.7 °C and 7.1 °C respectively (Isaacs and Mears, 1977; Clark, 1996, 2004).

Mean annual rainfall at Lucas Heights was 1013 mm for the period 1958–2008 (BOM, 2009). Monthly average rainfall varies between 52.5 mm in September and 112 mm in March.

Many of the significant rainfall events in the Sydney region result from coastal low-pressure systems transporting moisture from the Tasman Sea. Alternating dry and wet periods are common and often linked to wider climatic events such as El Niño, La Niña and may be modulated by the Indian Ocean Dipole (Ummenhofer et al., 2009). Since 1958 based on yearly rainfall deviation from the average, it is possible to differentiate three wet periods from 1958 to 1962, 1971–1977 and 1983–1991, alternating with dry periods from 1963 to 1970, 1978–1982, 1991–2007, the years 2007–2008 were relatively wet in contrast to the drought of the previous seven years. The annual average evaporation is ~1600 mm.

2.3. Neighbouring sites and local history

The neighbouring areas around the LFBG site have had diverse land uses including; disposal of liquid chemical waste, solid human waste, and three municipal landfill sites (Fig. 1B, C). All of these activities impose constraints on the future utilisation of the area and have the potential to affect the nearby LFBG groundwater hydrologically and hydrogeochemically. Approximately 400 m NW of LFBG, the Industrial Waste Collection Pty Ltd ("IWC") Liquid Waste Depot was used for the disposal of liquid and solid toxic wastes from 1969 to 1980 (Coffey, 1991). Directly adjacent to LFBG and to the E–NE, an extensive area of land was used for the disposal of solid human waste or "night soil" from 1947 to the early 1970s (Coffey, 1991). Also bordering the site along the west, a ~6 ha area known as Harringtons Quarry was excavated to a depth of ~6 m below the surface between the mid 1950s and 1984 (Coffey, 1991). In 1987 the quarry was filled with municipal waste after the closure of the Lucas Heights I landfill 1.5 km to the NE and before the opening of its replacement Lucas Heights II landfill 0.5 km to the SW of LFBG. A leachate drainage and pump-out system is in operation at this site.

2.4. Waste production, burial methods and timeline

A total of 79 trenches were used for waste disposal in the period 1960–1968 (Fig. 1C). Trenches were typically 25 m long, 0.6 m wide and 3 m deep and spaced at intervals of 2.7 m (AAEC, 1985). The waste was covered by about a metre of overburden and has been top-dressed as necessary, in part to compensate for the effects of settling.

The wastes consisted of solidified and solar-dried sludge from the AAEC effluent treatment plant, contaminated laboratory consumables and equipment, and waste packages consigned from other organisations such as universities and other government departments. The amounts of radioactivity in various operational classifications were recorded and these records provide a reasonable basis for estimating the total amounts of radionuclides disposed. This paper considers tritium disposal at the site. The tritium amount and form disposed of are unknown and it was rarely listed in the detailed disposal records. However, the waste also consisted of other radionuclides, such as fission and activation products, with the total amount disposed of being approximately 150 GBq (AAEC, 1985) including gram amounts of plutonium, ^{233}U and ^{235}U and larger amounts of ^{238}U, ^{232}Th as well as several hundred kg of beryllium.

2.5. Hydrogeology

Local hydrogeology affecting LFBG is characterised by a perched aquifer separated from the main Hawkesbury Sandstone by shale and siltstone lenses. These confining materials inhibit direct downward flow of water to the Hawkesbury Sandstone below. The Hawkesbury Sandstone has been interpreted as a layered aquifer system with groundwater occurring in discrete horizons with occasional vertical connection. Groundwater movement is interpreted to be variable with both primary granular flow and secondary flow along fractures (Grey and Ross, 2003). A conceptual model of LFBG hydrogeology is shown in Fig. 2.

Hydrographs measured in 1977 suggested that recharge of the perched aquifer was from precipitation over the burial ground with an effective runoff coefficient of 20% and a soil porosity of 10% (Isaacs and Mears, 1977). Groundwater discharge to the surface (“seepage”) observed to the north and south of the LFBG trenches (around MB13 and BH10 in Fig. 1C) following rainfall provides evidence that shallow subsurface flow pathways may be important in the transport of water and contaminants away from the site. Water from the shale–sandstone interface is also observed to emerge at the limits of the shale outcrop (AAEC, 1985) southwest of LFBG.

The various land uses of the surrounding areas, in particular Harringtons Quarry, have included excavation, burial of municipal waste, and leachate extraction, which would have had an effect on the site hydrological regime. In the 1970s there was extensive quarrying of shale adjacent to the western boundary of the LFBG (Harringtons Quarry, Fig. 1). Topographic maps from the mid 1970s, before quarrying ceased, show that the depth of excavation extended to the top of the Hawkesbury Sandstone and averaged ~6 m (AAEC, 1972). This is hypothesised to have changed the water table in the quarry area from conditions similar to that observed within the fenced perimeter of the LFBG site, to a frequently lowered water table today responding to leachate drainage and pumping. The removal of the confining layer has created a relatively transmissive pathway for movement of landfill contaminants not captured by the leachate system into the underlying Hawkesbury Sandstone aquifer, which has an effective porosity of 50% (Coffey, 1991).

3. Field and laboratory methods

3.1. Borehole construction

Several generations of observation bores are present on the LFBG site (Supplementary Data Figure S1 and Table S2). The earliest bores, numbered “BH” or “OS”, were established in the 1960s prior to or during waste disposal operations, and were part of a more extensive series of 24 boreholes which also covered the night soil area (AAEC, 1966). These are un-screened open bores partially lined with asbestos cement, or in the case of BHF, lined with PVC. The “D” series, established in the mid 1970s, are PVC lined boreholes mostly 3 m but up to 6 m in depth, screened from the surface and mostly located around the trench area (Isaacs and Mears, 1977). The “MB” series established in 1988 covers the fenced perimeter and also provides one key borehole (MB16) located near the centre of the trench area. The MB boreholes consist of slotted PVC pipe extending from ~0.5 m to various depths between 4 and 7 m, with most terminating within the shale materials above the sandstone. Bores CW, P1d and P2d are deeper boreholes that are screened within the underlying Hawkesbury Sandstone down to 12, 22 and 36 m respectively. P1s is the only borehole screened entirely within the unweathered shale layer. The more recent “P” and “CW” series were developed according to Australian borehole construction guidelines (LWBC, 2003).

Near-surface suction samplers were installed adjacent to BH10, MB13 and OS2 in August 2007, where surface seepage has been observed. The porous cup of each 50 cm diameter sampler was buried to a depth of ~0.25–0.3 m and backfilled with silica flour paste around the cup, then local clay and bentonite to the surface.

3.2. Water sampling methods

Tritium data is available from 1975 to the present. Prior to 2001, sampling equipment comprised of a centrifugal pump actioned with a petrol motor. This method was replaced by submergible piston-operated Bennet pumps, which are driven by compressed nitrogen and capable of sampling water from deep boreholes. The present sampling equipment incorporates a 12 V centrifugal pump having a low flow rate capability. Regardless of the pumping equipment all boreholes were purged of 3 bore volumes, or until dry, to enable fresh groundwater to infiltrate the bore. An unfiltered bulk 5-L sample for radiological testing is collected from each monitoring bore after the sampling container has been pre-rinsed with the sampled groundwater. Since August 2007, 0.45 μm filtered samples were also collected.

Soil water samples were collected from the suction samplers by first purging the sampler, then creating a vacuum of 0.6 bar. The samplers were left for ~24 h to allow the vacuum to draw soil water into the sampler through the porous cup, then the sample was collected using a peristaltic pump. Sample volume was a function of the soil moisture and varied from 0 to 0.4 L.

Rainwater samples were collected weekly since July 2004 at the ANSTO Meteorological Station located 180 m east of HIFAR, which was the main source of airborne tritium until it was shut down in 2007.

Unfiltered landfill leachate samples were collected either directly from a pump located in a sump at Harringtons Quarry or from the influent streams to the Lucas Heights Landfill leachate treatment plant.

3.3. Tritium analysis

Groundwater, soil water and leachate samples were prepared for liquid scintillation counting using either distillation (up to 2007) or Eichrom™ Tritium columns (from 2008 onwards). Until 1992 samples were distilled directly. The distillation method currently used follows the International Organization for Standardization method 9698 (ISO, 1989). This technique involves adding sodium carbonate and sodium thiosulphate prior to distillation.

The Eichrom™ Tritium column contains three extraction resins: the Diphonix™ resin removes cations; the AG 1X8 resin removes anions; and the XAD-7 resin removes organically bound ^{14}C and ^{3}H as well as other organic molecules (Eichrom, 1996). Initially, the storage solution in each column was allowed to drain before being washed with 10 mL of Milli-Q water. Approximately 25 mL of each sample was filtered through a 0.22 μm membrane filter and loaded onto a column. Leachate samples were treated with nitric acid and hydrogen peroxide to neutralise the pH and decompose organic compounds prior to being loaded onto the column. The first 5 mL of column eluate was discarded and the remaining 20 mL was collected.

Both the distillation and Eichrom sample preparation techniques could underestimate tritium activity if there is a significant portion of tritium that is bound to a mobile organic compound. However, the two methods have been compared and found to be statistically equivalent for transpirate samples taken from the LFBG (Twining et al., 2009).

Five millilitres of each purified sample was transferred to a HDPE LSC vial and 11 mL of Ultima Gold (XR or LLT) scintillation cocktail was added. Samples were dark equilibrated before being counted on either a Packard TriCarb 2700TR or 2900TR liquid scintillation analyser, along with a background and three standards prepared using the Amersham tritium standard TRY64. Each sample, standard and background was counted for 6 cycles of 20 min.

Different liquid scintillation counters, scintillation cocktails and sample to scintillation cocktail mass ratios have been used since beginning of tritium monitoring. Instrumental precision and the MDA (minimum detectable activity) have varied over the last 33 years of monitoring. In general, earlier samples reported an MDA of either 40 or 250 Bq/L and results below this level were not reported. From 1997 to the present the MDA ranges from 10 to 20 Bq/L.

3.4. Groundwater elevation

Standing water levels were not systematically recorded. However, some direct and indirect information is available. Isaacs and Mears (1977) recorded standing water levels during June–December 1974 for all BH series boreholes. Since November 2003 standing water level data has been collected prior to groundwater sampling. From July 2007 dedicated Odyssey pressure and temperature loggers have provided regular and systematic data.

4. Results

4.1. Tritium in groundwater

Between 1975 and 2008 more than 1000 groundwater samples were analysed for tritium with 36.7% being reported as less than the MDA. A statistical summary of the data both in its raw format and decay corrected to the time of disposal (taken as 1 July 1965) is given in Table 1. These data are listed in full in Table S3 of the Supplementary Data and are compiled from a large number of ANSTO Effluent Monitoring Reports and other studies (Isaacs and Mears, 1977; Ellis, 1977; AAEC/ANSTO Environmental Survey reports listed in Supplementary Data Table S1). Over that period of monitoring the maximum tritium activity was 3.9×10^5 Bq/L in a sample from borehole D10 collected in 1975 (Fig. 1C). In general, tritium activity decreases with distance from the trenches, and radioactive decay and dilution have resulted in a significant reduction in activity over time. Of the most recently gathered samples, the highest activity of 1929 Bq/L occurred in a sample from borehole BH10. The tritium results at LFBG are generally several orders of magnitude higher than tritium from shallow groundwater within Sydney Basin Hawkesbury Sandstone, with typical activities of ~2 tritium units (<1 Bq/L).

4.2. Other tritium data

Tritium concentrations have been measured in several types of samples at LFBG, apart from groundwaters, including soil waters, leachates from the landfills, rainfall and surface waters. All these types of data are useful to interpret the patterns of groundwater tritium distributions at LFBG and briefly outlined in this section.

Tritium activities in soil water collected from three suction samplers indicate that tritium levels appear to increase in shallow (~0.3 m) soil water during extended dry periods, however no simple correlation with rainfall is apparent (see Supplementary Data Table S4). The average tritium values are 96, 60 and 190 Bq/L for soil water sampler locations Lys1, Lys2 and Lys3 respectively.

Thirteen leachate samples from the Lucas Heights I and II landfills and from Harringtons Quarry were collected from November 2008 to January 2009 and analysed for tritium. These were supplemented by 12 Harringtons Quarry leachate samples collected and analysed between 1989 and 1992 (see Supplementary Data Table S5). Leachate from Lucas Heights I landfill (which operated from the late 1960s to 1986) had tritium activities averaging 139 Bq/L, and Lucas Heights II landfill (which commenced operation in 1987) had an average of 418 Bq/L. Over a similar period Harringtons Quarry leachate averaged 306 Bq/L, compared with a slightly higher average of 365 Bq/L in the early 1990s.

Tritium was measured in 175 rainfall samples collected between July 2004 and October 2008 at ANSTO (see ANSTO Environmental Survey reports, 2005, 2006, 2008 in Supplementary Data Table S1). Of these, 134 results were lower than the MDA (median MDA = 14 Bq/L, these were assumed to be 0.5 × MDA in calculations). The maximum tritium activity was 491 Bq/L and the rainfall amount weighted mean was 14 Bq/L.

Isaacs and Mears (1977) measured tritium in runoff in the drain near OS1 following rainfall and reported an average value of 111 Bq/L. Whilst tritium in surface water at LFBG has not been systematically measured during this study, two surface seepage samples and three creek samples were analysed: ponded seepage water near BH10 on 24 June 08 had 33 ± 13 Bq/L; seepage water near MB22 sampled on 6 August 08 had 53 ± 11 Bq/L. The creek downstream of the drain monitored by Isaacs and Mears was sampled on 8 October 08 after 7 mm of rainfall and had 23 ± 8 Bq/L of tritium, however samples collected on 15 December 08 and 17 December 08 following 56 mm of rainfall two days prior were <MDA (MDA = 18.7 Bq/L).

4.3. Groundwater elevation

Groundwater hydrographs show the varying response to rainfall in different borehole locations and constructions, and at different depths within the aquifer (Fig. 3). The muted and delayed responses of P1s and P1d are typical of boreholes screened below the topsoil/clay zone. In contrast, the MB series boreholes, which are screened to near surface, exhibit a rapid response to rainfall, with the hydrograph peak commonly rising to near the ground surface at the

Table 1
Statistical summary of tritium data (Bq/L) from 1975 to 2008.

	Average	Std deviation	First quartile	Median	Third quartile	Maximum	Number of results[a]	Number reported as <MDA
Raw data	7.6×10^3	3.0×10^4	1.3×10^2	7.6×10^2	3.6×10^4	3.9×10^5	637	369
Decay corrected	2.1×10^4	5.6×10^4	1.1×10^3	5.4×10^3	1.7×10^4	7.0×10^5	637	369

[a] Number of results not reported as <MDA.

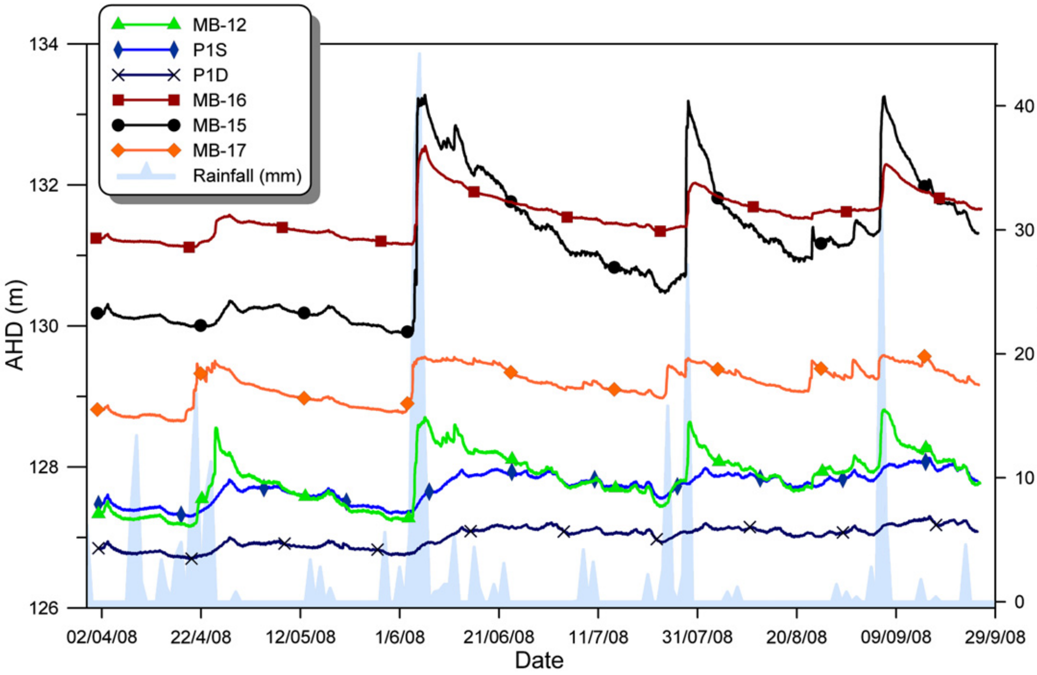

Fig. 3. Groundwater elevation and daily rainfall from April to September 2008. Well screened boreholes, P1s and P1d show gradual small responses to rainfall, whereas the shallow fully screened bores exhibit rapid response showing their level of connection with the surface. This demonstrates the degree of dilution that samples from these boreholes may be subject to.

peak of infiltration. This suggests that following sufficient rainfall, a shallow perched body of subsurface water can form (not sufficient to be considered an aquifer) independent of the underlying shale and sandstone aquifer. A sharp decline in the water table following the peak as seen at MB15 (and also at MB11, MB14 and MB18) suggests there is rapid drainage of water though the topsoil layer following a rain event, either down slope, or westwards to Haringtons Quarry where the leachate pumping system artificially lowers the water table. MB17 and MB13 show a response typical of boreholes in seepage zones. Their pre-event water tables are much closer to the surface compared with other boreholes, and following rainfall often exhibit a delay before receding. Whilst the hydraulic head of most shallow boreholes appears to respond to major and minor rainfall events, borehole MB16 appears to respond only to major events. This suggests influence of the nearby trenches that may provide storage and thus dampen event response compared to undisturbed locations.

4.4. Hydraulic conductivity

Recent slug-test measurements of saturated hydraulic conductivity (K_{sat}) have been performed at the site and analysed using methodology outlined by Amoozegar and Warrick (1986) for fully screened boreholes and Everts and Kanwar (1993) for the PS and CW series boreholes. The K_{sat} results are given in Supplementary Data Table S2. In the shallow BH/OS and MB series boreholes terminating in or above the shale layer, K_{sat} values ranged from 2.8 to 74 mm/day which is comparable to values reported by Isaacs and Mears (1977). These values are, however, at least two orders of magnitude higher than the range expected for consolidated shale sediments (Domenico and Schwartz, 1990). As these boreholes are essentially slotted for their entire length, the K_{sat} values are, therefore, more likely to be representative of the overlying weathered soil profile. The boreholes to the east in the night soil area (MB19–21) have consistently lower K_{sat} values than those in undisturbed ground at LFBG.

Hydraulic conductivity values measured in the P series boreholes CW and P1d, screened in the underlying Hawkesbury Sandstone, were generally higher than those measured in the shallow boreholes and are representative of this type of geological material. Although borehole P1s terminates in the shale, the high hydraulic conductivity measured in this borehole suggests that there is some connection to the confined aquifer in the Hawkesbury Sandstone, and this is also supported by continuous groundwater elevation data.

5. Discussion

5.1. Tritium sources and characterisation

Three potential sources of tritium in local groundwaters have been identified and their likely contribution to the levels of tritium measured in LFBG groundwater considered. These sources are atmospheric deposition through rainout of both locally and regionally derived tritium, tritium in waste buried at LFBG, and tritium in leachate from surrounding municipal landfills.

5.1.1. Atmospheric deposition

Tritium occurs in groundwater at LFBG, in part, as a result of atmospheric deposition. The atmospheric contributions originate partly from global deposition from natural and anthropogenic sources. At its peak in 1962, general tritium fallout over the Australian continent had a weighted average activity of 7.96 Bq/L, with that value declining to 0.49 Bq/L in 1985 (Calf, 1988). Present-day levels are even lower and considered to be consistent with natural background levels (Tadros et al., 2004), and are generally from one to four orders of magnitude lower than levels currently observed in LFBG groundwater. Additional atmospheric deposition of tritium is locally derived, associated with emissions from the Lucas Heights reactor (HIFAR), and has been measured at 14 Bq/L (rainfall weighted average from 2004 to 2007) 1.6 km from LFBG. These local rainwater activities are orders of magnitude lower than typical levels in the groundwater impacted by LFBG waste.

However, contributions from HIFAR could explain the above-background levels at boreholes in locations where direct impacts from trench wastes are thought to be unlikely when considering groundwater flow.

The distribution of tritium, and trends over time, indicate the primary source of tritium in groundwater within the LFBG boundaries is the waste buried within the trenches. The LFBG waste disposal records do not enable the precise source activity or disposal location to be definitively determined. Samples from the borehole at the centre of the trenched area (MB16) registered the highest sustained concentrations over time from 24,120 Bq/L in 1988 when first monitored, declining to present activities of 1378 Bq/L largely due to radioactive decay (Supplementary Data Table S3). In addition, the data also present a distribution pattern consistent with a dispersing plume having the trenches as its source. Furthermore, the persistence of the tritium at the centre of the trenched area for more than forty years (>3 half lives) suggests there is ongoing release of tritium from the waste.

5.1.2. *LFBG waste*

A calculation was performed to determine if the total amount of tritium in the groundwater at LFBG is consistent with waste amounts available for disposal in the 1960s. The assumptions used in this calculation are: 1) the effective porosities of the shale/clay, sandstone and buried waste are 0.1, 0.5 and 0.5 respectively (Coffey, 1991; Isaacs and Mears, 1977); 2) the depth of the shale/clay layer is 9 m (from AAEC, 1966); 3) the plume extends to the bounds of contours and to a depth of 40 m, (this assumption is based on tritium being detected at depth in P2d); 4) the concentration is uniform vertically over that depth range; and 5) the HIFAR D_2O tritium activity in 1965 was 3.8 mCi/mL (Morgan, 1966). Based on the March 2008 contours (Fig. 4), the total tritium activity present in groundwater at the site was estimated to be ~400 GBq. Decay corrected to 1 July 1965 this would have amounted to 4 TBq at the time of disposal. This amount could have been waste from laboratory or tracer studies using tritiated water, or is the equivalent of 30 L of HIFAR heavy water, which could potentially have been present in decontamination wastes. This is a relatively small volume relative to total trench volume. It is also a relatively small amount of tritium which may have been accumulated as waste from laboratory or field tracer studies at the AAEC or other institutions.

5.1.3. *Municipal landfill leachate*

An additional potential tritium source is the adjacent municipal landfill (Harringtons Quarry). Tritium is known to be present in municipal landfill leachate. A number of landfill leachate studies in the UK and USA have reported tritium concentrations in municipal landfill leachate to be routinely above background (Hackley et al., 1996; Mutch and Mahony, 2008; Robinson and Gronow, 1996; Hicks et al., 2000), with maximums up to 7098 Bq/L and averages of 797 Bq/L. The likely tritium sources in landfill include a range of household goods such as luminous watches and clocks (<280 MBq/device), luminous telephone dials (<1 GBq/device), and gaseous tritium light sources (<7.5 GBq/device). Tritium levels ranging from 134 to 760 Bq/L have been measured in leachate from landfills in the vicinity of LFBG (Harringtons Quarry and Lucas Heights I and II, Supplementary Data Table S5). The ranges of average tritium levels measured in the three landfills correspond well to the expected decay of the tritium based on the average age of deposited waste, suggesting that continuing leaching of tritium of the waste is occurring. The current leachate capture system at Harringtons Quarry is estimated to extract an average tritium amount of 3 MBq/d or 1 GBq/y (based on an average tritium leachate concentration of 306 Bq/L and an average volume pumped of 9725 L/d). While most of the leachate from Harringtons Quarry may be intersected by the leachate drainage and pumping system, the base and sides of the landfill are unlined, therefore some leachate may migrate horizontally to the LFBG, and some may migrate downward directly into the Hawkesbury Sandstone providing another source of tritium to the local groundwater system.

In conclusion, the patterns of groundwater tritium show that two sources are major contributors, namely the radioactive waste emplaced at LFBG and leachate from the nearby Harringtons Quarry landfill. While the LFBG is the major contributor in the vicinity of the LFBG trenches, the landfill leachate contributes a generally elevated background and needs to be taken into account in modelling the system. Leachate from the Lucas Heights I and II landfills is unlikely to be detected at LFBG but may be present in groundwater in the wider area. Atmospheric deposition is a minor contributor to tritium concentrations in groundwaters.

5.2. *Tritium migration and response to climate*

Tritium activities in groundwater at LFBG vary considerably over time, with major changes correlated to wet and dry climatic periods (Fig. 5). To better understand tritium variations, these results are interpreted with reference to the cumulative monthly rainfall deficit. This is defined as the cumulative sum of measured monthly rainfall minus average monthly rainfall (commencing at July 1978). This provides a local indicator of trends in average rainfall values. Declining trends indicate dryer conditions while rising trends indicate wetter conditions. For this analysis, tritium data were decay-corrected to 1965 to allow changes due to dilution or dispersion to be separated from radioactive decay.

The MB16 borehole is located near the centre of the trench source area and therefore its tritium levels provide an indicator of changes in release rates from the waste as a response to rainfall and infiltration. MB16 is also located in the area of trenches with the highest elevation and is therefore only influenced by infiltration during wet-dry cycles, not by surface runoff or seepage. Data from MB16 indicate a general decline in tritium activity since 1988. However, this evolution has irregularities that can be divided broadly into three stages. The stage from 1988 to 1995 is characterised by high and relatively constant tritium activities (~70,000 Bq/L decay corrected to 1965) with a small response to the prevailing wet climate conditions at the time (Fig. 5). These high values suggest a consistently elevated source, or sources, of tritium leaching from the waste trenches. A second stage from approximately 1995 to 2002 coincides with the onset of a dry period. During this period tritium variations are extreme and do not correlate clearly with rainfall deficit. This pattern suggests that the most consistently elevated source(s) had decreased and the adjacent levels in the monitoring boreholes were more subject to dilution following rainfall events. The third stage from 2002 to 2006 correlates with an extended period of drought; during this period, tritium activities increased consistently due to the prevailing drought conditions with lower groundwater levels and marked reduction in dilution of the existing tritium (Fig. 5). A return to average rainfall conditions in 2007 was accompanied by significant reduction in tritium activities as the system became dilution dominated again.

Limited data exist on vertical movement of tritium through the clay and shale near the trenched area. Isaacs and Mears (1977) reported that tritium levels were higher in the deeper of two piezometer pairs (3 m at D15/D18 and 6 m at D15a/D18a) suggesting that downward vertical movement of tritium was occurring at the trenches. The only other multilevel data available are from MB12/P1s/P1d located in the drainage line SE of the trenches and MB18/CW/P2d located at the NW corner adjacent to Harringtons

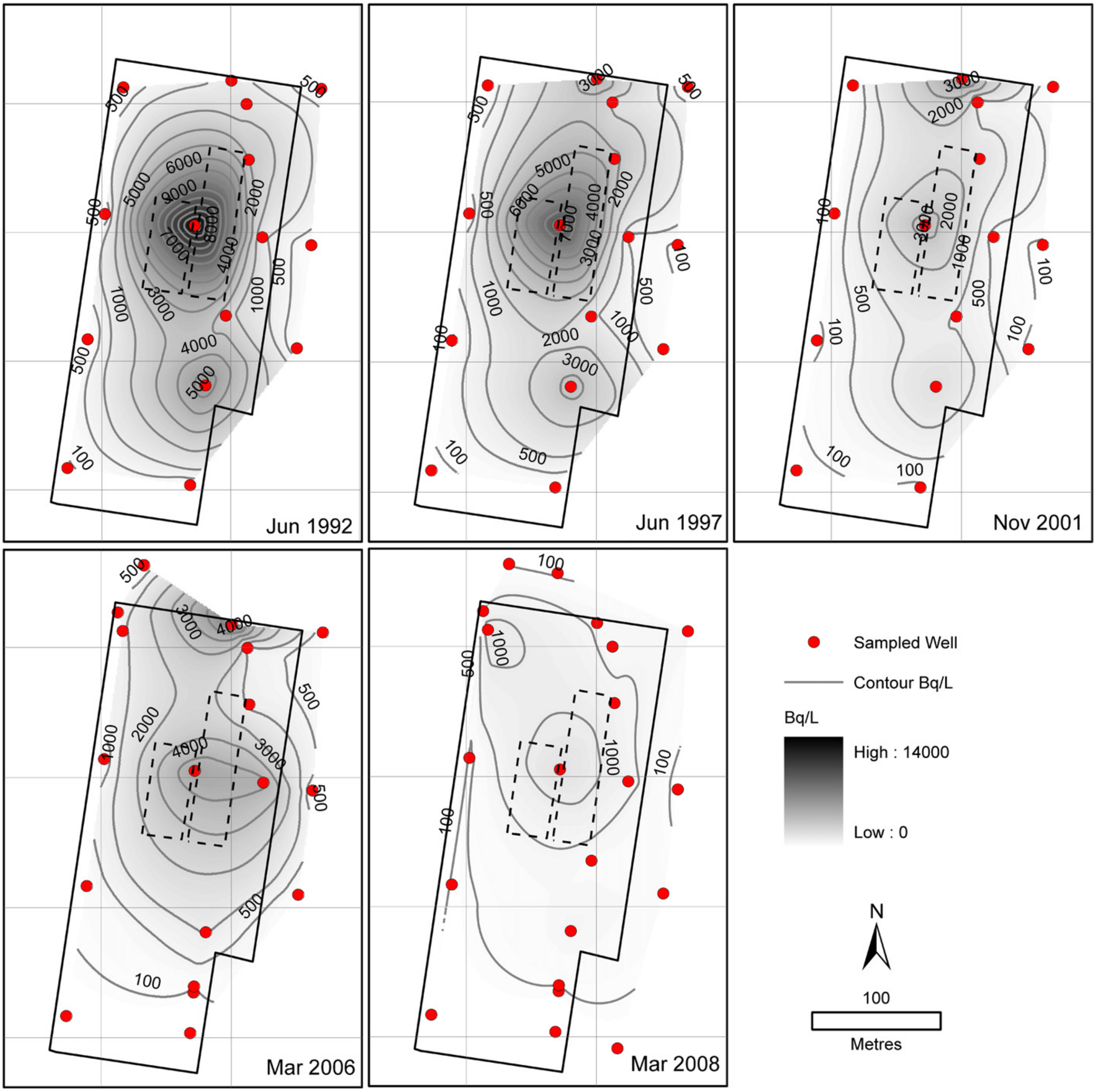

Fig. 4. Tritium contours (not decay corrected) for June 1992, June 1997, November 2001, March 2006 and March 2008 show the progressive decrease in activity over time. Two other features are evident in this time series, firstly the increase in activity in 2006 following extended drought, and secondly, an increase in tritium to the north of the trenches.

Quarry. At both of these sites there is a downward hydraulic gradient.

Hydrograph and tritium data from MB12, P1s and P1d (Fig. 3) provide some evidence of rapid shallow subsurface flow. P1d is the deepest borehole of the three, screened fully within the sandstone aquifer, P1s is screened fully within the shale layer, and MB12 is screened from the shale up to the surface clay layer. The MB12 hydrograph follows P1s trends closely during dry weather, but exhibits higher elevation increases in response to rainfall. This suggests that infiltration from shallow flow pathways dominates the borehole response and that the clay layer acts as an aquitard. Isaacs and Mears (1977) proposed that shallow subsurface flow may be a significant pathway for contaminant movement away from the trenches and data presented here support this. Tritium was present in soil water and surface water seepage at locations down gradient of the trenches. In addition, the borehole hydrographs show that the perched water table comes to near the surface during rainfall events providing a pathway for horizontal movement of tritium and exchange with surface water.

To illustrate the changes in tritium distribution across the site, tritium concentrations (Supplementary Data Table S3) were interpolated and contoured for five sampling times between 1992 and 2008 (Fig. 4). During the first snapshot (Jun 92) the tritium plume was concentrated over the MB16 area, over time there has been a gradual dispersion and migration northwards (Nov 01, Mar 06). The increases in tritium activity observed in BH10 and BHF (Fig. 5)

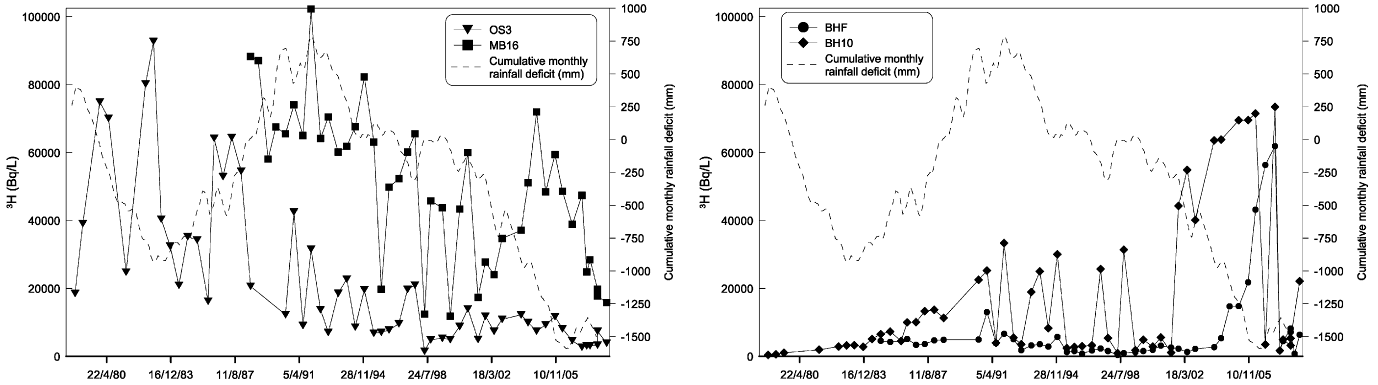

Fig. 5. Decay corrected tritium activities at OS3, MB16, BHF and BH10 from July 1978 to 2008 compared to cumulative monthly rainfall deficit ([Monthly rainfall – average monthly rainfall] + preceding period balance). MB16 and OS3 are located within or directly adjacent to the trenches and show a gradual decline in the peak activities at or near the source. BHF and BH10 are located along the flow path and show an increase over the period peaking at the height of the 2002–6 drought.

during the latest drought period were accompanied by smaller but definite increases in MB14, 15 and 18 along the western boundary and MB19 outside the NE boundary, despite that fact that these areas were not previously considered to be affected by the contaminant plume (Figs. 4 and 5). Prior to this period tritium levels in the MB series samples were normally less than minimum detectable activities.

The observed increases during drought may be the result of continuous release rates from the source trenches with dilution influences occurring during tritium movement. Tritium can move away from the trenches along three possible pathways: a) vertical migration through the shale into the sandstone aquifer; b) lateral flow in or above the shale layer; and c) through shallow perched groundwater or interflow following rain events. During wet periods the interflow pathway transports tritium laterally near the surface, where it is further diluted as it flows downhill following surface contours; this water dilutes any deeper plume during sampling from boreholes that are screened to the surface such as the MB and BH series. During dry periods, when the water table is lower, this contaminant pathway is less active and samples are more likely to reflect contaminant levels in the underlying shale aquifer which may have higher tritium activities. The longer time series of BH10 (Fig. 5) in particular shows larger variations in tritium concentration that could support this hypothesis.

An additional potential mechanism that may explain the increases in tritium concentrations, even during periods of rainfall deficit, is the arrival of the rising limb of the breakthrough curve of the tritium plume in boreholes away from the source trenches. The evidence for this is a gradual increase in tritium to the north (BH10, Fig. 5), sustained even during lower and declining activities at the source (MB16 and OS3, Fig. 5). The same pattern is observed east in BHF but not south in MB13 indicating variable source release or transport rates. Along the western boundary, the increases in tritium in MB14, 15 and 18 also show this pattern but at levels that are similar to or lower than those seen in Harringtons Quarry leachate so it is also possible that shallow tritium along the western boundary may be sourced from there. Harringtons Quarry leachate pump-out data from Jan 2005 to September 2008 indicate that while large rainfall events do result in increased leachate quantities, leachate generation is continuous even during extended dry periods providing a potential source of contamination to surrounding areas.

The short time series of tritium data from groundwater in the Hawkesbury Sandstone do not show a strong correlation to wet-dry cycles. Borehole P1d has higher tritium activities than P2d and may show a subtle response with higher activities during dryer periods and the influence of LFBG tritium. However, P2d showed smaller and constant activities that could be related to inputs from landfill waste in Harringtons Quarry.

6. Conclusion

The tritium data at LFBG reflect the response and transport of a conservative tracer during the ~45-year period since wastes were buried in shallow trenches in the 1960s. The pattern of tritium distribution suggests that the predominant source of the tritium plume at the LFBG is the waste materials in the LFBG trenches. However, additional sources contributing to above-background tritium levels in local area groundwater include the adjacent Harringtons Quarry landfill as well as potentially other landfill sources. The existence of multiple sources requires a complex conceptual model when interpreting tritium data from boreholes which are more distant from the LFBG, particularly those located in the boundary area between the LFBG and Harringtons Quarry. Future work could include trace element analysis designed to distinguish between LFBG and Harringtons Quarry leachate sources.

The persistence of elevated tritium levels immediately adjacent to the trenches after more than 40 years since disposal indicate ongoing source release from wastes. Further, release rates and dilution rates appear to respond to wet and dry rainfall periods with elevation of activity concentrations occurring during extended dry periods. Data to date suggest the elevated tritium levels are representative of shallow groundwater that is relatively undiluted by infiltration of rainwater, whilst the temporarily decreased levels represent more diluted conditions following rainfall events. In addition to the short-term fluctuations, data also indicate more gradual increases and decreases in tritium levels at various locations near the trenches consistent with general movement of a tritium plume away from the source area. Data from sampling locations north of the trenches indicated increasing trends of tritium levels (a gradual breakthrough curve) during extended dry periods, documenting that plume movement is not arrested by drought.

Whilst the period of peak activity appears to have passed, less conservative contaminants, including isotopes of strontium, plutonium, and uranium among others are still present in the waste at LFBG. The tritium data presented here provide insights into the hydrological processes and potential direction and pathways for future migration of contaminants that should be considered in designing further studies or in management of the site. Beyond the conceptual model presented here, the tritium data may also be used as a conservative tracer for future validation of a groundwater and contaminant transport model of the site and to guide further site investigations.

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Appendix. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jenvrad.2010.05.009.

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