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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

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ABSTRACT

The results of a 21 month sampling program measuring tritium in tree transpirate with respect to local sources are reported. The aim was to assess the potential of tree transpirate to indicate the presence of sub-surface seepage plumes.

Transpirate gathered from trees near low-level nuclear waste disposal trenches contained activity concentrations of ^{3}H that were significantly higher (up to $\sim$700 Bq L^{-1}) than local background levels (0–10 Bq L^{-1}). The effects of the waste source declined rapidly with distance to be at background levels within 10s of metres. A research reactor 1.6 km south of the site contributed significant ($p < 0.01$) local fallout ^{3}H but its influence did not reach as far as the disposal trenches.

The elevated ^{3}H levels in transpirate were, however, substantially lower than groundwater concentrations measured across the site (ranging from 0 to 91% with a median of 2%). Temporal patterns of tree transpirate ^{3}H, together with local meteorological observations, indicate that soil water within the active root zones comprised a mixture of seepage and rainfall infiltration. The degree of mixing was variable given that the soil water activity concentrations were heterogeneous at a scale equivalent to the effective rooting volume of the trees. In addition, water taken up by roots was not well mixed within the trees. Based on correlation modelling, net rainfall less evaporation (a surrogate for infiltration) over a period of from 2 to 3 weeks prior to sampling seems to be the optimum predictor of transpirate ^{3}H variability for any sampled tree at this site.

The results demonstrate successful use of ^{3}H in transpirate from trees to indicate the presence and general extent of sub-surface contamination at a low-level nuclear waste site.

1. Introduction and aim

The Little Forest Burial Ground (LFBG) is a legacy, near-surface, waste disposal site located at Lucas Heights near the old HIgh Flux Australian Reactor (HIFAR, Fig. 1) in southern Sydney. Both the LFBG (via groundwater) and HIFAR (via atmospheric dispersal) are potential sources of tritium (^{3}H) to the nearby environment. Disposal operations at the LFBG took place from 1960 to 1968 (Isaacs and Mears, 1977) and ^{3}H comprised part of the waste. Since then, the site has been regularly maintained and routine monitoring of groundwater, grass and high volume air sampling, as reported in annual ANSTO Environmental Monitoring reports (e.g. Hoffmann et al., 2008), has continued to the present day. Recently initiated studies at the site have been more intensive than the routine work and have been directed towards better understanding, and modelling of, the behaviour of radioactive materials under Australian conditions as well as to assess management options for the site. Aspects of the improved modelling will hopefully be transferrable to other Australian sites, such as the proposed national nuclear waste storage facility under consideration.

At any waste site, one of the key issues is the potential hazard to humans or other species that may arise due to the movement of radionuclides through the environment to potential receptors. It is therefore necessary to understand the environmental pathways which may lead to such exposures, which usually include the possibility of movement of radionuclides in groundwaters through the vicinity of the site. It was believed that trees at the LFBG had the potential to intersect any sub-surface dispersion plumes from the buried waste, thereby providing a more expansive spatial coverage than the existing set of bores at the site and allowing sampling over

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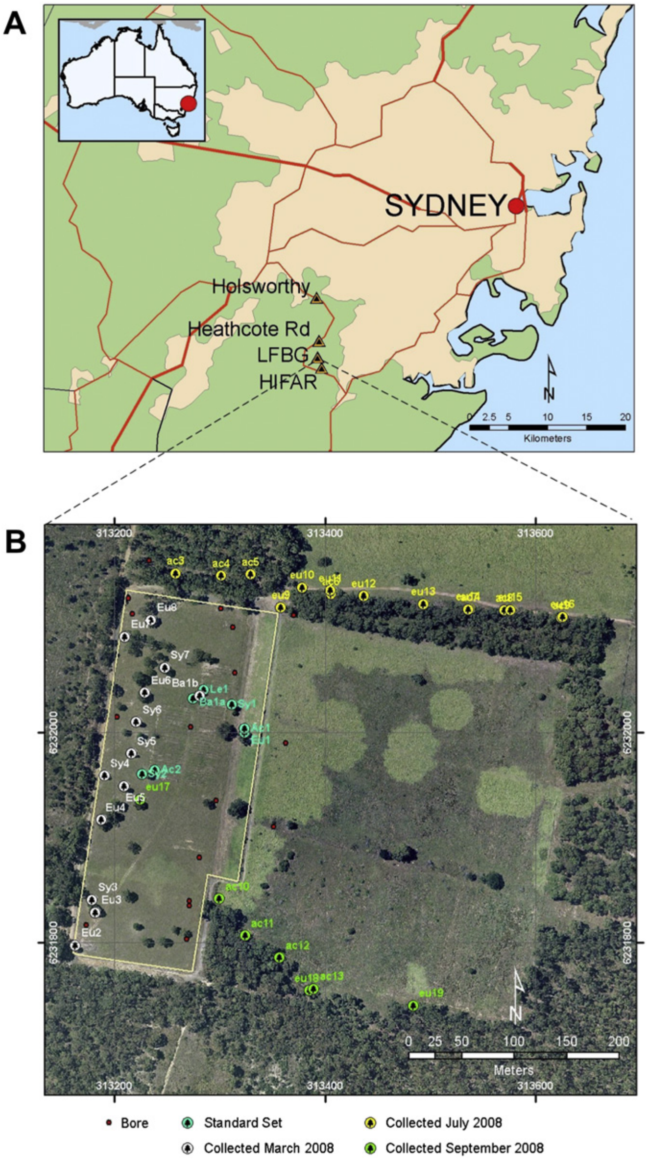

Fig. 1. The Little Forest Burial Ground and nearby HIFAR research reactor. A, Location map and sampling sites for long, NS transect; B, Aerial view of LFBG showing locations of sampled trees comprising standard set and nearby transects.

the waste without otherwise disturbing it. Tritiated water, being a conservative tracer of such a plume, would be taken up by plants and transpired. The soil–root–leaf pathway was identified as the major transport mechanism for ^{3}H from contaminated groundwater by Evenden et al. (1998). Thus, this radioecological study was instigated in 2007 to assess that potential.

Since that time, there has also been an increasing emphasis placed on the need to undertake environmental dose assessment of nuclear activities (e.g. ICRP 108, 2008). This paper will report on the findings of a 21 month survey of transpirate ^{3}H over, and adjacent to, the trenches and with respect to the other local ^{3}H source, HIFAR. Whilst ^{3}H itself is not of undue concern from a radiological perspective, particularly at the levels measured in this investigation, these results will be used to identify the trees which have substantial access to any waste seepage plume. Those trees will be targeted for additional tissue analyses for other radionuclides to assess their relative transport rates in the plume as well as their bioaccumulation rates for dose assessment modelling purposes.

2. Materials and methods

2.1. Site description

At the LFBG, a variety of low-level nuclear waste materials were disposed of in a series of 77 unlined trenches ~3 m deep, 25 m long and spaced 2.7 m apart. As each trench was filled, it was re-covered with topsoil to a depth of ~1.0 m. There was a total of 1675 m^3 of waste, comprising ~150 GBq of activity and including 1730 kg of beryllium (Isaacs and Mears, 1977). The locations of trenches are still evident from minor surface subsidence at the present day (Fig. 1b).

The local geology, described by Hughes et al. (2010), consists of topsoil and clay to a depth of 1.5–2 m, which is derived from weathered shale to a depth of approximately 6–8 m. The shale overlies the Hawkesbury Sandstone which extends to a depth of around 200 m. Surrounding the trenches, a perched saturated zone exists near the top of the weathered shale at ~3 m. This saturated zone is hypothesised to be fed by leachate from the LFBG trenches. Below this level other perched saturated zones have been observed, with the main aquifer occurring in the Hawkesbury Sandstone.

2.2. Sampling

A variety of approaches were assessed to determine the best methods for sampling and analysis of ^{3}H in transpirate. More detailed descriptions of those techniques are available in Twining et al. (2009). The following is a synopsis of the optimal methodology applied over the period.

2.2.1. Biota selection

Seven accessible trees and shrubs over, or adjacent to, the trenches were identified as being most likely to intersect any subsurface seepage plumes. These plants, comprising a gum tree (*Eucalyptus paniculata*; Eu 1), two similar *Acacia* sp (Ac 1, *Acacia obtusifolia* and Ac 2, *Acacia longifolia longifolia*), two turpentine trees (*Syncarpia glomulifera*; Sy 1 and 2), a *Leptospermum polygalifolium* shrub (Le 1) and a *Banksia serrata* tree (Ba 1) are identified in Fig. 1b and are hereafter referred to as the standard set. Typical rooting depths for these species are not well established, particularly for this site; however the following values are estimates provided from the scientific literature. In schlerophyllous scrub and forest in south eastern Australia the range of maximum rooting depths for *Banksia* spp and a *Leptospermum* sp. is 2.3–2.5 m (Canadell et al., 1996, citing Specht and Rayson, 1957). McColl (1969) reported an average rooting depth of just 1.2 m for *E. paniculata* based on observations of soil profiles in a coastal forest south of Sydney. No information on the rooting depths of the *Acacia* spp nor of the *Syncarpia* sp. in this study is otherwise available, although Canadell et al. (1996, Fig. 2 therein) provided an estimated mean maximum rooting depth for trees of 6.9 ± 1.2 m and for shrubs of 5.0 ± 0.9. Average rooting depths will be somewhat less than the maxima and it should be recognised that local soil conditions will also constrain plant growth.

Initial samples of transpirate were collected in Jul 2007. Subsequent sets of samples using the same trees and as far as possible the same branches, were collected approximately every three months until Mar 2009. In Oct 2007, samples were collected from more branches on the trees identified as Ba 1, Sy 1 and Ac 2 (Fig. 1b) to better estimate variability within these trees. In addition, three transects of approximately 400 m length located adjacent to the trenches were sampled on or about 13 Mar 2008 (West); 24 Jun 2008 (North) and 30 Sep 2008 (South East) (Fig. 1b). A fourth, much longer transect (~15 km) beginning at the HIFAR reactor which is approximately 1.6 km south of the LFBG, then running northward

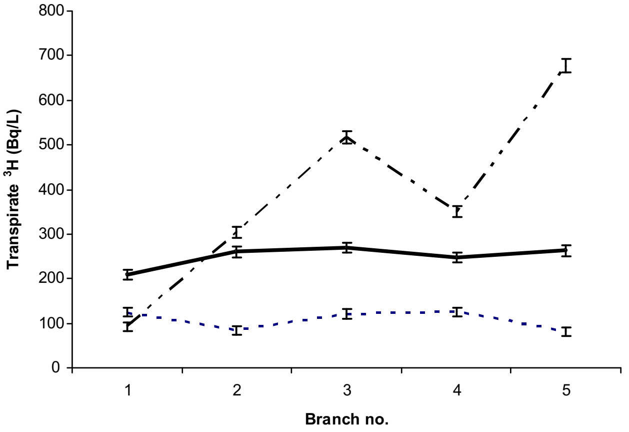

Fig. 2. Tritium activity concentrations in transpirate collected from 5 branches on each of 3 trees [Ac 1 (dotted), Ba 1 (dashed) and Sy 1 (solid line) refer to Section 2.2.1 for sample details] on 4 October 2007. Analytical errors (±1 s.d.) are also shown.

through the LFBG and beyond (Fig. 1a) was sampled on 30 Sep 2008 and 23 Mar 2009. Plants sampled in these transects comprised *Eucalyptus*; *Syncarpia* and *Acacia* spp.

2.2.2. Collection of transpirate

The leaves at the distal end of a limb (in the case of trees or large shrubs) or limbs (in the case of smaller shrubs) were enclosed in a large, dry, clean, clear PVC bag with the open end being taped closed around the branch (or branches) using adhesive cloth tape. The bag was typically placed on a north-facing (sun-exposed) limb, on sunny days after dew evaporation, for four or more hours.

The condensate in the bag was collected with new, 50 mL disposable syringes. The samples were filtered *in situ* using dual stage, 1.0 and 0.45 μm polypropylene membrane cartridges, into clean, dry, HDPE plastic bottles. Volumes from 0 to >250 mL (typical range 50–150 mL) were collected over the entire period with smaller volumes collected on cooler, more overcast days. The filtered samples were returned to ANSTO and placed into a cool room to await further processing.

2.2.3. Sample processing

From the filtered transpirates, 25 mL aliquots were processed for ^{3}H analysis using Eichrom tritium columns (Eichrom, 1996) to remove dissolved organics (including organically-bound ^{3}H) and ions. After treatment, 5 mL of each sample was added to translucent, HDPE scintillation vials with 11 mL of Ultima Gold uLLT liquid scintillant cocktail and the vials were sealed for counting. A similar procedure was used to prepare standards and blanks to be included in each counting run.

2.3. Analysis

Samples, with blanks (distilled 'dead' water acquired from an aquifer with no bomb pulse ^{3}H) and standards (prepared using Amersham TRY64, batch 133, 1 Nov 2002), were counted on a Packard Tricarb 2900TR LSC using up to 31 × 20 min cycles over an energy range from 2 to 18.6 keV. Detection efficiencies were approximately 25% with a detection limit of $\sim$10 Bq L^{-1}. Quench correction used instrument T_{SIE} values and a correction curve derived from a set of 10 standard samples quenched with increasing amounts of food dye. The ^{3}H activity in each sample was determined using total counts, after background subtraction and accounting for within-run efficiency (standards) and quenching (T_{SIE}). Measurement uncertainties quoted in this study include counting, weighing and standard activity uncertainties.

2.4. Additional data

Meteorology observations form part of the routine monitoring program carried out at ANSTO. A range of variables are collected at frequencies up to every 15 min from the Lucas Heights meteorological station (Clark, 2003) approximately 1.6 km south from the LFBG. Daily averages of temperature, rainfall and pan evaporation were used to help interpret the variability in transpirate ^{3}H over time.

Similarly, routine water samples have been regularly collected and analysed for a range of contaminants, including ^{3}H, from boreholes across the LFBG and rainfall from the meteorological station (Hughes et al., 2010). In addition, water samples were gathered from series of shallow (3–5 m) core holes immediately after drilling near the trenches in Aug 2009.These results were used to assist interpretation of the spatial variability observed in the transpirate ^{3}H data.

3. Results and discussion

The full set of results for ^{3}H in transpirate at LFBG is available from the records at ANSTO via the first author. The results reported hereafter are subsets or averages relevant to the hypotheses being assessed in each case. All further references to ^{3}H in transpirate, soil or groundwater at this site refer to non-organically-bound ^{3}H in water.

3.1. Within-tree variability

Results for ^{3}H in transpirate collected on 4 Oct 2007 from 5 different branches on each of 3 trees (Ac 2; Ba 1 and Sy 1) are shown in Fig. 2. The measurement uncertainties are generally within the symbol of each datum. Hence, it can readily be seen that there were significant ($p < 0.01$) differences in ^{3}H being measured from different branches within any tree. The absolute differences between trees will be discussed below. Variability was greater in Ac 2 than in Ba 1 which was similar to Sy 1. The trees increased in size in that same order, so it may be that this pattern was related to rooting depth. However, rooting depth was not recorded to confirm this.

It is clear that the trees were all exhibiting different ^{3}H levels in different branches. This observation points to two related factors. Firstly, the roots were accessing different concentrations of ^{3}H in the soil (i.e. heterogeneous sources) and secondly, there was limited mixing occurring within the trees once water was taken up by the roots.

Regarding small scale variability at field sites, Amano and Garten (1991) encountered very heterogeneous soil water ^{3}H concentrations at a site adjacent to the Oak River National Laboratory in the USA. They relied on integration within the plant to assess average soil water concentrations for their modelling. Combining several leaf samples to provide a soil water value for their modelling may have been sufficient to overcome the variability identified in that study. Modellers within the IAEA BIOMASS Program also agreed that ^{3}H concentrations in soils at sites with sub-surface contamination can vary sharply over short distances (IAEA, 2003). From that, good predictions of ^{3}H transport required sophisticated models and detailed information on meteorological conditions and soil characteristics unless the models were generalised over larger areas.

The limited mixing of ^{3}H within tree water streams was not unexpected and similar disparities had been observed for tritiated water uptake in another Australian species (Twining, 1994). Xylem tissues are the main conductive vessels carrying water from roots to leaves for photosynthesis. Studies carried out in the rainforests of the Amazon (Jordan and Kline, 1977) and in coniferous areas of north-western USA (Kline et al., 1976) conclusively showed that the rate of transpiration in either environment was directly proportional to the sapwood cross-sectional area, and hence to the amount of xylem tissue present. These vascular tissues need to be highly conductive and yet with reduced lateral porosity to allow for water to be drawn vertically over substantial distances. In support of reduced horizontal mixing, Mathew et al. (1988) injected tritiated water into a number of holes bored into the base of a 2.5 m palm tree and then observed the appearance of ^{3}H in the leaves at the crown by sampling leaflets over time (the water was collected by vacuum freeze-drying and hence did not include any organically-bound ^{3}H). They found that the ^{3}H peak arrival varied from ~5.5–9 days in different leaves, almost twice the time from one branch to the other. This indicated that some flow paths were more tortuous than others but also that those paths were essentially discrete.

Evidence for some horizontal mixing does exist, however. Kline et al. (1976) undertook a similar study in which tritiated water was injected into holes near the base of Douglas fir trees. The vertical and horizontal movement of ^{3}H after 10 days was assessed by taking cores into the pith of the tree at 20, 30, 40 and 50 m heights above the injection point. As expected, these showed the most rapid vertical movement occurred towards the inner parts of the cortex (sapwood) where the xylem predominates. Most of the ^{3}H was at 40 m but a peak was starting to become apparent at 50 m. However some was also detected in the pith (heartwood). The pattern in this case suggested that ^{3}H moved horizontally out of the xylem and into the pith during the passage of the peak activity but then moved back into the xylem after the peak had passed. There was little or no evidence of vertical movement within the pith. None of these studies measured organically-bound ^{3}H.

3.2. Temporal variability in transpirate tritium activity concentrations

Variability in average transpirate ^{3}H over the entire sampling period for the standard set of trees is shown in Fig. 3. The values for each time period were mostly significantly different ($p < 0.01$) from the previous datum for each branch, although the degree of change was variable. The marked differences on 4 Oct 2007 were, to some degree, due to the sampling of multiple branches on some trees that day, as discussed earlier. Apart from that excursion, most of the trees retained a similar degree of activity concentration in transpirate over time despite the significant differences between each sample. For example, Sy 1 always had higher average ^{3}H activity concentrations than did Le 1, which in turn always had more than Ba 1 and similarly for Ac 1. This pointed to heterogeneity in the spatial distribution of ^{3}H in the soil profile, as indicated previously by the heterogeneity observed between branches and as will be discussed further below. It also indicated that the soil ^{3}H levels at any one location remained generally consistent over time. At a similar site to the LFBG, near the Oak Ridge National Laboratory Waste Storage Area, Amano and Garten (1991, citing Amano et al. 1987) reported that the change in soil ^{3}H concentrations was fairly slow, as reflected in our data.

There was also a tendency for the trees to follow a similar trend from one date to the next, with most trees either increasing or decreasing their average activity concentration simultaneously. This suggested that environmental factors were having some influence across the site, the most obvious being rainfall which will tend to dilute the ^{3}H activity concentrations in the surface soils as well as in the trenches (assuming that the trenches are the major source of ^{3}H in this location). Amano and Garten (1992) showed surface dilution leading to reduced ^{3}H values in the top 10 cm of the soil profile at the Oak Ridge site over both summer and winter compared to consistently higher activity concentrations for the next 70 cm. At LFBG, infiltration into the clay soils below the topsoil zone is believed to be slow. However, evidence suggests that the poorly capped trenches provide a preferential flow pathway allowing rainfall to dilute the ^{3}H in the trenches. The resulting saturated layer, observed at a depth of 2.5–3.5 m, may therefore have ^{3}H activity concentrations that also vary in response to rainfall.

The effect of rainfall is not only to dilute the surface soil ^{3}H. Plants can also respond to changing water potential by altering their active root zone, i.e. the depth at which they obtain their water, to access water at energetically more favourable levels (Guswa, 2008 and citations within). This is a physiological response to access water that requires less effort to adsorb and transpire. Over an annual cycle, Stringer et al. (1989) studied oaks on a hillside in Kentucky that was also affected by lateral groundwater flow of ^{3}H from the nearby low-level waste repository at Maxey Flats. In spring, the surface soils, from the top of the hill slope to lower down, had a good water content of about 0.4 m^3 m^{-3} due to snow melt; xylem water potentials in trees were all low (around −0.2 MPa); and foliage ^{3}H was also relatively low (at about 5×10^3 Bq L^{-1}). By well into summer, the surface soils had dried by about a factor of 2 and the trees at the top of the hill slope had increased their water potential up to about −1.75 MPa, indicating greater stress. These plants had also retained their earlier foliage ^{3}H concentrations. The trees at the bottom of the slope, however, had only mildly increased their water potential (to about −0.35 MPa) but their foliage ^{3}H had increased dramatically to more than 1×10^6 Bq L^{-1}. The trees had shifted their water source to the deeper, but relatively more accessible, seepage from the Maxey Flats site which was unavailable to the plants higher up the hill slope. The plants at LFBG may be similarly switching their water source in response to rainfall inputs and thereby temporarily changing their access to any potential sub-surface seepage plumes from the trenches.

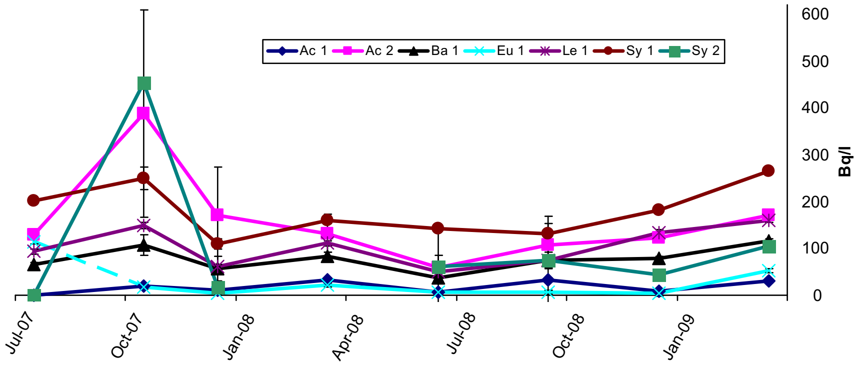

Fig. 3. Average ^{3}H activity concentration in transpirate from each tree (see Section 2.2.1 for tree details) from the standard set on each sampling date. The dotted line for Eu 1 indicates that a different branch was selected and sampled after July 2007.

To test this hypothesis, daily data of rainfall less pan evaporation measured at Lucas Heights were used to estimate a local value for the cumulative rainfall deficit as a surrogate for potential infiltration. The effects of daily temperature ranges over the period of the study were also assessed. These were compared with the percentage shift in transpirate ^{3}H activity concentration from one sample to the next for each branch over the entire sampling period. Percentages were used rather than the absolute differences to normalise samples to a common baseline. It should be noted that some data were excluded from these analyses, as follows: Two branches (Le 1.2 and Sy 2.2) had too few points with successive ^{3}H measures to allow for accurate regression analysis; branch Eu 1.1 had consistently low activity concentrations at less than twice the background and hence was prone to excessive variability due to counting uncertainty; and one branch (Ac 2.1) died soon after final sampling and hence could be considered to have potentially abnormal water use patterns.

Initial observations showed no direct relationship between the shift in transpirate ^{3}H and daily temperatures, rainfall events or cumulative rainfall deficit, however there was some indication that a leading average rainfall deficit (representing potential infiltration), estimated over some days or weeks, may be having an effect. Modellers within the IAEA BIOMASS Program Tritium Working Group also agreed that the balance between rainfall and evaporation, together with mechanical dispersion processes in soil, were key factors in the transport of ^{3}H through the unsaturated layer above a contaminated zone (IAEA, 2003). Fig. 4 a and b show the trends for shifts in transpirate ^{3}H concentrations for larger and smaller trees, respectively, using an arbitrary lag in the potential infiltration of 14 days. The correlations are not significant ($r^2 = 0.18$ and 0.46 respectively), however a consistent trend was observable. To determine an optimum lag period, the transpirate ^{3}H activity concentrations were correlated with net rainfall deficit over daily time steps from 1 to 90 days using a modelling approach described in Venables and Ripley (2002). Fig. 5 shows the results of the correlation modelling. Values around zero imply poor or no correlation between the number of days over which approximate infiltration was calculated and the measured shift in transpirate ^{3}H

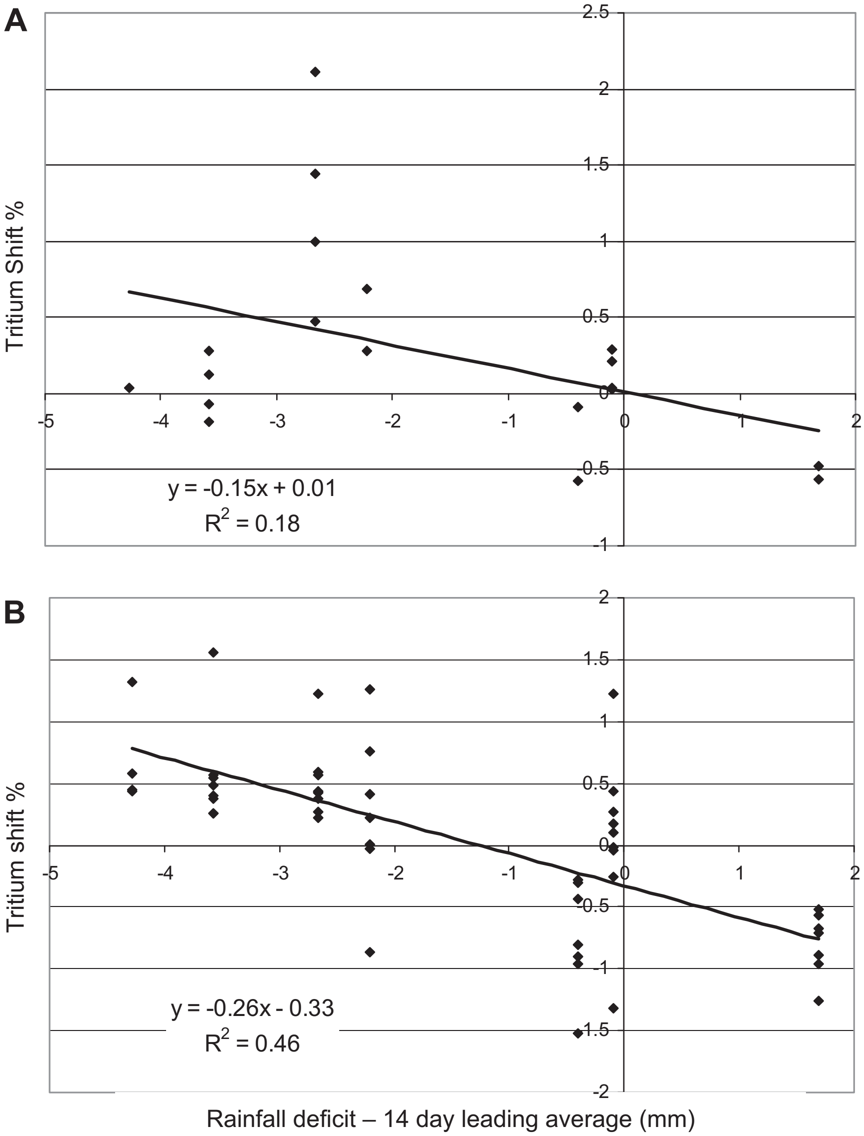

Fig. 4. Correlations of the % shift in transpirate tritium activity concentration in individual branches between successive samples at LFBG with the cumulative rainfall minus pan evaporation at ANSTO (1.6 km S) for the preceding 14 days. A, larger trees: *Eucalyptus paniculata* and *Syncarpia glomulifera*; B, smaller trees and shrubs; *Acacia longifolia longifolia* and *Acacia obtusifolia*, *Banksia serrata* and *Leptospermum polygalifolium*.

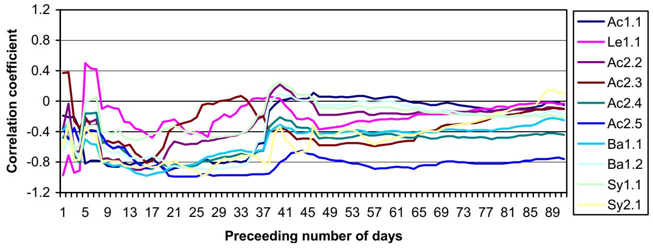

Fig. 5. Correlations for the % shift in tritium activity concentrations between sequential samples for individual branches at LFBG with the rainfall deficit at ANSTO (1.6 km S) over the number of days prior to sampling.

activity concentration. Negative values imply an inverse relationship (i.e. ^{3}H activity concentrations decrease with increasing rainfall) and the converse is true for positive values.

It is apparent from Fig. 5 that there was considerable noise in the data. A simple correlation using rainfall deficit alone did not fully explain temporal shifts in ^{3}H activity concentration in transpirate at LFBG, remembering also that the meteorological station was approximately 1.6 km distant from the sampled trees. Measuring additional parameters at the time of sampling, such as soil moisture content, air pressure, relative humidity etc. may have provided more modelling power to assist in evaluation. However, some points of consistency were discerned.

Firstly, most of the correlations were negative. This supports the hypothesis that rainfall infiltration into surface soil was diluting the activity concentration of ^{3}H in the active root zones. Secondly, the correlation curves tend to flatten off after about 40 cumulative days, suggesting that additional information after that time did not add anything to improve the interpretation.

Thirdly, there was considerable variation in correlation between branches in the period up to about 10 days. This implied that short-term variability in rainfall and evaporation had an imprecise effect on transpirate ^{3}H shift. Experimental studies with sunflower and tobacco plants showed that equilibrium between the ^{3}H in the solution to which their roots were exposed and the stems and petioles of mature leaves was reached within half a day (Raney and Vaadia, 1965). Citrus seedlings, being slightly larger plants at 1–2 years old, required up to 40 h to achieve equilibrium between a ^{3}H enriched nutrient solution bathing the roots and their leaves (Mantell et al., 1979). It follows that the larger trees sampled in this study will take somewhat longer times for such transfer to take place. All the literature sited in Section 3.1 (above) also indicated that there was a delay in mass transport of water within the tree in that it took from days to weeks between uptake from roots and subsequent transpiration. In addition, there is the time required for the infiltrating water to reach and mix in the root zone before any uptake can occur. Hence, it is not unexpected that recent, short-term, rainfall patterns will not be consistently reflected in the transpirate.

Lastly from Fig. 5, there was a period from approximately two to three weeks where most of the correlations were more strongly negative. This implied that a lag period in this range was optimal and, hence, the data in Fig. 4 were reasonable estimates for this site.

Table 1
Tritium activity concentrations in tree transpirate collected over or adjacent to the trenches at LFBG, or in nearby transects.

Sample set	Average ± S.E. (Bq L^{-1})	*n*	Range (Bq L^{-1})	Genera sampled
Standard	116 ± 11	100	0–667	See text
North East Transect	2 ± 1	13	0–10	*Eucalyptus* & *Acacia*
West Transect	12 ± 3	11	4–29	*Eucalyptus* & *Syncarpia*
South East Transect	23 ± 7	7	0–52	*Eucalyptus* & *Acacia*

3.3. *Spatial variability in transpirate tritium activity concentrations*

3.3.1. *Differences between the standard set and nearby transects*

Average transpirate ^{3}H activity concentrations for the standard set of trees and for the nearby transects indicated in Fig. 1b are presented in Table 1.

It is quite clear that significantly higher average activity concentrations were registered over the trenches than in the nearby transects. The transect to the north and east was at background levels. There was no evidence of any near-surface seepage plume in that direction. The transect to the west was also low but slightly higher on average than the northern transect. The LFBG is bordered by a disused landfill site in that direction which exhibited leachate ^{3}H levels ranging from 240 to 415 Bq L^{-1} in late 2008 (Hughes et al., 2010). It is possible than the minimal transpired ^{3}H levels detected could have been the result of seepage from the landfill into the LFBG instead of seepage from the trenches in this case. The transect to the south east follows a natural drainage line (Fig. 1b) where elevated ^{3}H levels have been found in surface runoff (Hughes et al., 2010). Hence, the slightly higher values again in that case may well have been associated with surface expression of a near-surface seepage plume or runoff from the LFBG trenches. However, the values were still much lower than those detected in the standard set and hence the seepage, if it existed, was negligible or mixed with lower activity waters. It should be noted that in none of the transects was there any significant difference in transpirate ^{3}H activity concentration between the different species that were sampled.

At the time of sampling, there was another potential nearby source of ^{3}H, the HIFAR research reactor approximately 1.6 km to the south (Fig. 1a), that may have contributed some activity by localised atmospheric transport, particularly to the south east transect. This is particularly so as it is reported that atmospheric ^{3}H uptake by plants tends to be the most common exposure pathway (Boyer et al., 2009). Whilst ^{3}H in rainfall at the Lucas Heights meteorological station averaged 14 Bq L^{-1} over the period 2004–2008 (Hughes et al., 2010), during the period of transpirate sampling the rainfall weighted average ^{3}H was 2.5 Bq L^{-1} following the final shutdown of HIFAR in the first quarter of 2007. To assess the atmospheric contribution to transpirate, a transect from HIFAR to the north through the LFBG was sampled twice. The results are shown in Fig. 6.

The local effect of HIFAR was clearly evident with transpirate levels well in excess of those observed in rainfall; however its

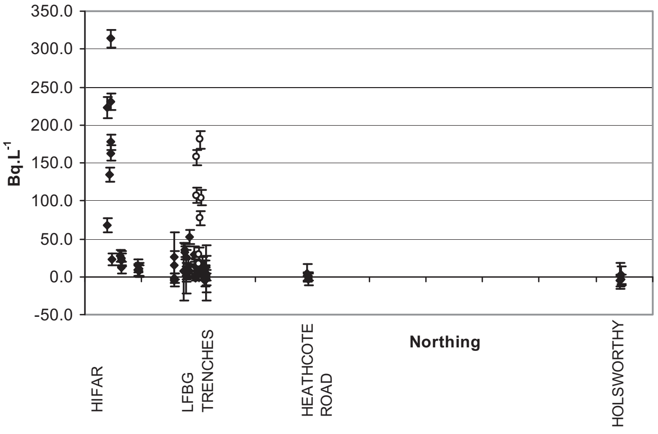

Fig. 6. Tritium activity concentrations in transpirate samples collected on two days along a transect from HIFAR through the LBFG and further north. The standard tree set is indicated by open symbols.

influence declined to background before the LFBG was reached. Background levels were re-established to the north of the trenches before the Heathcote Road samples. Therefore the standard set of samples clearly showed the effect of ^{3}H sourced from the trenches. In addition, samples near the trenches showed an increase above background which implied some movement, albeit small, into the wider environment at LFBG from either the LFBG trenches or the adjacent Harringtons Quarry landfill. This was reinforced by recent data acquired as a consequence of drilling a series of shallow core holes with depths of 3–5 m surrounding and between the trenches, including one transect of 6 core holes perpendicular to the trenches at distances of ~1 to ~15 m. The core extraction work indicated a shallow, perched, saturated zone within 2–3.5 m of the ground surface in the area of the trenches and corresponding approximately with the base of the burial trenches. Samples were gathered from seepage waters that collected in the core holes over the 24-h period immediately after drilling in Aug 2009. The ^{3}H concentrations in the transect waters were up to ~2.7 × 10^4 Bq L^{-1} closest to the trenches. However, they declined quickly to <5 × 10^3 Bq L^{-1} within 15 m.

3.3.2. Groundwater interactions

Four sets of measurements of groundwater ^{3}H made in 1975 and 1976 by Isaacs and Mears (1977) showed that the highest levels (>2 × 10^5 Bq L^{-1}) occurred in samples taken from shallow boreholes located between the two sets of trenches, and this was confirmed during the Aug 2009 core hole sampling. Tritiated water has been measured in groundwater samples from LFBG twice yearly since 1975 as part of the routine monitoring program and an analysis of the results has been reported by Hughes et al. (2010). The monitoring indicated that ^{3}H in groundwater has been slowly dispersing from the trenched area, particularly towards the north and south, broadly following the surface gradient. The current ^{3}H maxima are from 3 to 4 × 10^4 Bq L^{-1} as indicated by the Aug 2009 sampling of core holes.

The potential for capillary rise of ^{3}H contaminated groundwater in the clay soils of the LFBG would be in the order of 10 m (Holtz and Kovacs, 1981, Tables 6–1) allowing ^{3}H to be transported vertically up the soil profile during dry periods. This vertical transport process will mediate any potential tree interaction with the groundwater plume.

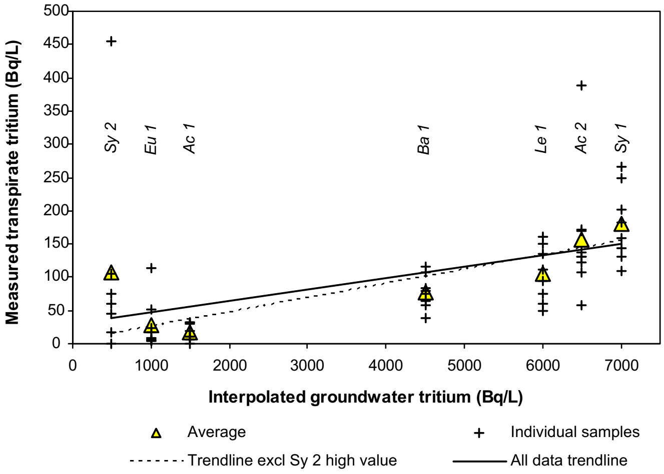

Fig. 7. Transpirate vs groundwater ^{3}H at LFBG. Groundwater values were estimated from interpolation of Aug–Nov 2009 core hole and groundwater samples. The relationship between transpirate (TR) and groundwater (GW) for all data is $H^3_{TR} = 0.017\ H^3_{GW} + 29.0$ ($r^2 = 0.24$). When the highest Sy 2 data point is removed the correlation improves and the intercept becomes closer to zero: $H^3_{TR} = 0.021\ H^3_{GW} + 4.9$ ($r^2 = 0.51$).

Given that the locations of the available groundwater boreholes do not correspond with those of the tree samples, groundwater ^{3}H values from the Aug 2009 core hole samples and selected Oct–Nov 2009 borehole samples were interpolated to estimate the groundwater ^{3}H concentration at the seven standard transpirate sampling locations. The interpolated groundwater values are plotted against the transpirate values in Fig. 7. Soil water samples collected using lysimeters to a depth of ~30 cm at three locations at the LFBG were reported to have ^{3}H levels ranging from 28 to 253 Bq L^{-1} (Hughes et al., 2010), similar to those found in transpirate in this study; however their locations were not directly comparable with the vegetation sampled in this study and were intentionally located in surface seepage zones.

The ^{3}H activity concentrations in the transpirate (and lysimeter) samples were substantially less than those measured in waters collected from boreholes across the LFBG site. Average transpirate concentrations from individual trees and sampling dates ranged from 0 to 91% of the estimated groundwater concentration, with an overall average of 4.5% and median of 2%. However, a number of factors were likely to influence the relationship between transpirate and groundwater, such as recent rainfall, depth to groundwater, rooting depth and the presence of preferential flow pathways. Similarly, there were simplifications made in interpolation of the groundwater at the transpirate sampling points including assumed linear variation in activity (unlikely to be true), ignoring differences in bore depth and bore construction, variation in standing water levels and poorly defined ^{3}H distribution in the trenches. Despite that, an overall positive correlation between groundwater and transpirate ^{3}H can be seen in Fig. 7 with an r^2 value of 0.24. *Syncarpia* 2, however, exhibited a much higher average transpirate to groundwater ratio, which may indicate that the groundwater concentration was underestimated at this location and/or that preferential flow pathways or root locations close to the trenches provided the tree roots access to more concentrated groundwater. When the highest data point from Sy 2 is excluded the r^2 increases to 0.51 and the intercept comes nearer to zero. Nonetheless, the observation that all of the average transpirate values were less than any interpolated groundwater value points to the fact that the soil water being accessed by the plants is a mixed source, likely to comprise rainwater diluted seepage. This is consistent with the observation, above, that the transpirate ^{3}H concentrations tended to respond inversely to rainfall intensity.

4. Conclusions

Transpired water from trees growing over, or adjacent to, the low-level nuclear waste disposal trenches at the LFBG contained activity concentrations of ^{3}H that were significantly higher (up to a factor of about 70 times) than local background levels. There was a correlation between proximity to likely seepage water plumes and ^{3}H activity concentration in transpired water.

The levels in transpirate were substantially lower than groundwater concentrations measured across the site (by a median factor of 50). The observed shifts in tree transpirate ^{3}H concentration between sampling periods, together with local meteorological observations, indicate that the soil water within the active root zones comprises a mixture of seepage and rainfall infiltration. The degree of mixing was variable given that the soil water activity concentrations were heterogeneous at a scale equivalent to the effective rooting volume of the trees. In addition, water taken up by roots was not well mixed within the trees.

Whilst the nearby HIFAR research reactor was a source of atmospheric ^{3}H and local fallout over the period of sampling, its influence reduced to background within the distance to the LFBG. However, the low-level waste at the LFBG had a slight, but measurable, effect on transpirate ^{3}H activity concentrations beyond the disposal trenches. Those effects declined rapidly with distance to be approaching, or at, background levels within 10s of metres from the trenches. It is possible that some ^{3}H contribution may be coming from an alternative low-level source, the adjacent landfill to the west of the trenches, but if so, it is minimal.

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