

Tritium Concentrations in Crops Fertilised with Contaminated Sewage Sludge

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ABSTRACT

The Food Standards Agency (FSA) let a contract to the Radiation Protection Division of the Health Protection Agency (RPD) to exploit the research opportunity afforded by discharges of tritium from a radiopharmaceutical facility to a waste water treatment works (WWTW) in Cardiff. Dried sludge pellets from the WWTW were used as a soil conditioner and the uptake of tritium from the treated soil into crops was investigated. The project was divided into two parts. In 2005 a pilot study was performed to determine whether uptake was measurable and decide whether tritium was being lost rapidly from the soil. This pilot study was outside the constraints of the Sludge (Use in Agriculture) Regulations. In 2006 the study was repeated without further addition of sewage sludge which provided conditions that did conform to the regulations. The tritium concentration in soil fell throughout the study. The changes in concentrations of tritium in soil over the growing seasons and the low concentrations measured in crops meant that it was not possible to quantify individual soil to crop transfer in terms of conventional concentration ratios. However it was possible to determine an aggregated transfer quotient relating the concentration in the edible part of the crop in Bq kg^{-1} fresh mass to the original amount of activity applied to the soil in Bq m^{-2} . For 2006 and for all of the crop - soil combinations studied, values of about 2×10^{-4} were derived. The observed values are likely to depend on factors such as temperature, rainfall and any artificial irrigation. Consequently, those derived in this study are not necessarily applicable in general radiological assessments. However, on the basis of these indicative values it would be reasonable to assume that the proportion of tritium transferred into crops from soil treated with sludge would be small.

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EXECUTIVE SUMMARY

Sewage sludge is being used increasingly as a soil conditioner on agricultural land. Authorised discharges of radionuclides into sewers can result in contamination of the sludge, so together these factors can provide a route by which radionuclides can enter the foodchain. Currently, a radiopharmaceutical facility in Cardiff discharges effluent into a new waste water treatment works, which in turn produces sludge pellets for use in agriculture. These pellets could therefore provide an opportunity to study uptake of radionuclides from treated land. Samples of these pellets were analysed and the results showed that concentrations of tritium were sufficient for such studies to be worthwhile. No other radionuclides were present in sufficient quantities to warrant further study.

The HPA Radiation Protection Division has a lysimeter facility that was established in 1983. This contains 3 diverse soil types – a sandy soil, a loam and a peat. The soils in these lysimeters were treated with a known amount of sludge pellets in 2005. At the same time, sets of smaller containers were filled with fresh samples of the same soils. To extend the range of soils being studied, a clay soil was also collected for use in the small containers. Known amounts of sludge pellets were added to some of the smaller containers; others were left untreated to check on ambient levels of tritium in the environment or whether any tritium being released into the atmosphere from the treated soils was significantly affecting the concentrations measured in nearby crops. The purpose of using the smaller containers was to extend the range of crops that could be studied and also to enable any losses of tritium in leachate, ie water draining out of the soil, to be monitored.

The use of sewage sludge on agricultural land is controlled by the Sludge (Use in Agriculture) Regulations, often referred to as the Safe Sludge Matrix. Under these Regulations, land that had been treated in 2005 could not be used for crops until 2006. In this study, crops were grown in 2005 to determine whether uptake was measurable. In addition, the changes in activity concentrations in soil and in leachate from some of the small containers were investigated. In 2006, further crops were grown in conformance with the regulations, while changes in activity concentrations in soil and drainage water were checked less frequently. A final set of soil samples was collected in 2007.

Concentrations of tritium in all soils decreased throughout the period of the study. In broad terms, about 80% of the tritium originally applied was lost over the two growing seasons (2005 and 2006), with the rate of loss being lower in 2006. The tritium that remained in the soil was mainly in an organic form, usually referred to as OBT (organically-bound tritium). In general, the OBT remained in the surface layer of the soil where it had originally been incorporated.

There was a marked loss of tritium via leachate from all of the soil types studied. Losses were higher during 2005. In this case the tritium was in the form of tritiated water. However, the amounts of activity in leachate were not sufficient to account for all of the losses of activity implied from the measurements in soil. When the activity transferred to crops was taken into account, it seems likely that tritium was also being

lost via transpiration from the soil or from the crops, or from a combination of the two. A more sophisticated experiment would be needed to quantify losses via these processes.

Concentrations of tritium were measurable in most crops grown in treated soil but were generally low. Results for most crops grown in untreated soils were generally near to or below the limit of detection, which indicated that cross-contamination via transpiration from nearby treated soils and crops was not significant. For all vegetable crops, the activity was mainly in the form of tritiated water. However, all of the crops had a high water content, and it was likely that concentrations of any OBT would have been below the limit of detection. Samples of grass generally contained higher amounts of OBT than vegetable crops. However, the concentrations of OBT were variable. Overall, the indications were that soil splash was an important contributor to the amounts of tritium observed in the grass. The implications for radiological assessments are that inadvertent ingestion of soil could be an important contributor to intakes of tritium by animals grazing on land that has been treated with sewage sludge.

The changes in activity concentrations in soil over the study period, coupled with the low concentrations measured in the crops, meant that it was not possible to quantify the uptake of tritium using the conventional transfer factor approach adopted for many other radionuclides. However, an aggregated transfer quotient was derived to relate the activity concentration in the edible part of the crop (in Bq per kilogramme fresh mass) to the amount of activity originally applied (in Bq per metre squared). The units for this quotient are therefore metres squared per kilogramme ($\text{m}^2 \text{kg}^{-1}$). For 2006, ie for crops grown in conformance with the Safe Sludge Matrix, a value of about $2 \cdot 10^{-4} \text{m}^2 \text{kg}^{-1}$ was obtained for all soil – crop combinations. The observed values are likely to depend on factors such as temperature, rainfall and any artificial irrigation. Consequently, those derived in this study are not necessarily applicable in general radiological assessments. However, on the basis of these indicative values it would be reasonable to assume that the proportion of tritium transferred into crops from treated soil would be small.

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CONTENTS

1	Introduction	1
2	Study design and initiation	2
	2.1 Starting materials	2
	2.2 Approach adopted	2
	2.3 Supporting analyses	4
3	Results and discussion	6
	3.1 Concentrations of tritium in soil	6
	3.2 Transport of tritium in the soil	10
	3.2.1 Migration down the soil column	10
	3.2.2 Transport out of the soil in leachate	10
	3.2.3 Potential importance of deposition in rainfall	12
	3.3 Transfer of tritium into vegetables	12
	3.4 Grass	16
4	Conclusions	17
5	Acknowledgements	19
6	References	20
	APPENDIX A	21
	Analytical results	21
	A1 Tritium in soil	21
	A2 Tritium in leachate and rainwater	30
	A3 Tritium in crops	32
	A4 Tritium in grass	38
	APPENDIX B	41
	Comparison with values in predictive models	41
	B1 Approach adopted	41
	B2 Conclusions	42
	B3 Reference	42

1 INTRODUCTION

The extent to which sewage sludge is used as a soil conditioner is increasing. Authorised discharges of radionuclides into sewers can result in contamination of the sludge, and so together these factors provide a pathway by which radionuclides can enter the foodchain. A review of published information, carried out by the National Radiological Protection Board (NRPB), indicated that specific data on the uptake of radionuclides from sludge amended land into crops was scarce. Current discharges from a radiopharmaceutical facility in Cardiff are presently routed to a new waste water treatment works, and have resulted in elevated concentrations of some radionuclides in sewage sludge pellets. There is therefore the potential to generate specific data on radionuclide uptake by crops grown in soil amended with sludge from this treatment works.

In 2004, the Food Standards Agency (FSA) let a contract to the NRPB to exploit this possible research opportunity. NRPB became the Radiation Protection Division of the Health Protection Agency (RPD) in April 2005. Provided that activity concentrations in the sludge were sufficient, the aim of the work was to make use of the RPD's existing lysimeter facility at Chilton to generate realistic uptake data for selected crops grown in three soil types. This was to be augmented by smaller scale studies on a wider range of crops that would generate relative values for uptake parameters, based on the same soils. The original aim was to combine these two sets of data to provide parameter values that could then be applied in the FSA's predictive models.

Currently, there is very considerable uncertainty about the rate at which tritium can be lost from soil, either via transpiration or via downward migration. This has important implications for situations involving the application of tritium-containing sludge to agricultural land because of the constraints of the Sludge (Use in Agriculture) Regulations (commonly referred to as the Safe Sludge Matrix). These regulations require that crops cannot be harvested from treated arable land until at least 10 months after the sludge has been applied. The aims of the first phase of this project, carried out during 2005, were to determine whether uptake was measurable in crops grown in treated soil and to investigate whether tritium was being lost rapidly from the soil. It was recognised that, in terms of crop production, this part of the study was outside the constraints of the Safe Sludge Matrix. The results of this phase of the work were then used to aid the design of the next phase of the work in 2006. The experiments in 2006 focused on uptake into plants under growing conditions that did conform with the Safe Sludge Matrix.

2 STUDY DESIGN AND INITIATION

2.1 Starting materials

About 300 kg of sewage sludge pellets were collected from the Cardiff East waste water treatment works on 30th November 2004. An initial analysis of the tritium content was performed at NRPB's Glasgow laboratory. In terms of mass as received, the average concentration measured was about 87,000 Bq kg⁻¹ of total tritium. Nearly all of the activity was in what was operationally-defined as an organically bound form. Concentrations of ¹⁴C and gamma-ray emitting radionuclides were also determined. Where these were detectable, the measured values were significantly below those for ³H and were not considered sufficient for crop uptake studies to be worthwhile. These radionuclides have not been considered further in this project.

The lysimeter facility at the RPD Chilton laboratory is set out in three pairs, with each pair containing a different soil type. The soil types employed are Hamble loam, Fifield sand and Adventurers peat. Each soil was collected from suitable field sites in 1983. Each lysimeter is 1.7 m in diameter and 0.5 m in depth.

To enable a larger range of crops and soil types to be studied, the existing lysimeters were supplemented by a series of smaller containers. Water cisterns of 20 l capacity were used with dimensions of 0.45 m * 0.3 m * 0.27 m (w*d*h). To provide the soils for these new containers, the locations originally used in 1983 were revisited and further samples were collected during the early spring of 2005. In addition, samples of a clay soil of the Denchworth series were also collected from a site that had been identified in 1983. Together, these four soils provide a suitable contrast in the types of agricultural soil found across the UK.

Drainage holes were drilled in each of the new containers and unique identifiers were assigned. A set of 18 containers was used for each type of soil. After each container had been filled, the soil was tilled and then allowed to settle. At about the same time the soils in the established lysimeters were dug over and allowed to settle.

2.2 Approach adopted

The overall scheme of container numbering, soil type, and the crops grown in 2005 and 2006 is shown in Table 1. Table 2 lists the varieties of each crop that were used. Each soil : crop combination involved 3 of the smaller containers. Two of these were treated with sludge pellets; the third remained untreated and was used as a control. This was considered important because of the possibility of transpiration, ie the transfer of tritium in the vapour phase between containers. This approach also provided a check that ambient levels of tritium in the environment were not affecting the results.

Once the soil had settled after tillage, the designated containers and the six lysimeters were amended with sewage sludge pellets at a rate corresponding to about 20 tonnes per hectare (2 kg m⁻²) which is equivalent to 1.7 10⁵ Bq m⁻² of tritium. This exceeded the maximum rate specified in the Safe Sludge Matrix, but was considered necessary

because the measured concentrations of tritium in the pellets were lower than had originally been expected. This higher rate had been agreed in advance with the FSA project officer. The pellets were incorporated into the top 100 mm of soil using standard gardening tools. The soils were mixed again 3 and 7 days after amendment.

For 2005, one important objective was to improve understanding of the behaviour of tritium in soil. For this reason, samples of soil were taken from the lysimeters throughout the growing season. The results will be discussed in detail later but the rate of change in the concentration of tritium in soil decreased through 2005. For this reason, in 2006 soil was only sampled at the beginning and end of the growing season. One soil sample of each soil type was also taken from the lysimeters in June 2007, to investigate the effects of a second winter. Each time that samples of soil were collected, aliquots of about 20 g were taken. The total amount of soil in the smaller containers was insufficient for this approach to be adopted without disturbing the crops. Consequently, soils in the smaller containers were sampled only before planting or sowing and then again when a crop was harvested. After collection, all soil samples were sieved through a mesh of approximately 3 mm in size. One aliquot was dried at 105°C to determine moisture content and a second aliquot stored at -20°C for tritium analysis. The smaller containers provided an opportunity to collect and analyse leachate, ie the liquid that had percolated through the soil and was drained away. One container of each soil type that had been sown with grass was used for this purpose.

Crops were sown or planted in late May 2005 for the first phase and in early May 2006 for the second phase. With the exception of the permanent crops (grass and strawberries), crops were rotated between containers over the two years. They were maintained according to good horticultural practice, watered as required and regularly fed with a proprietary liquid feed. Grass was cut to about 30 mm above the soil surface whenever it reached about 150 mm in height. All other crops were collected at maturity. All crops except grass were washed in tap water and inedible parts discarded. The entire sample of the edible part was then roughly chopped or shredded. One aliquot was dried at 105°C to determine moisture content and a second stored at -20°C for tritium analysis. Potatoes and carrots were not peeled and were analysed as complete tubers or roots. In 2005, an additional sample of one potato crop was taken from a lysimeter. This was used to determine the distribution of activity between peel and flesh. For several other radionuclides, the concentration in the peel is greater than in the flesh (Green et al, 1997), and such information is therefore valuable when specific radiological assessments are required. In 2005, the lysimeters were able to produce more than enough mature crop for the subsequent analyses, and so in 2006 a wider range of crops was grown in them, under conditions that more representative of those in the field.

A rainwater sampler of 300 mm diameter was installed a few metres away from the small containers and lysimeters in order to keep a check on any ambient levels of tritium in the general environment

Table 1 Container soil and crop assignment

Container Number and soil type					
Sandy	Clay	Loam	Peat	2005 Crop	2006 Crop
S1	C1	L1	P1	Dwarf French Beans 1	Potatoes 1
S2	C2	L2	P2	Dwarf French Beans 2	Potatoes 2
S3	C3	L3	P3	Dwarf French Beans Control	Potatoes Control
S4	C4	L4	P4	Cabbage 1	Carrot 1
S5	C5	L5	P5	Cabbage 2	Carrot 2
S6	C6	L6	P6	Cabbage Control	Carrot Control
S7	C7	L7	P7	Carrot 1	Dwarf French Beans 1
S8	C8	L8	P8	Carrot 2	Dwarf French Beans 2
S9	C9	L9	P9	Carrot Control	Dwarf French Beans Control
S10	C10	L10	P10	Grass 1 (collect leachate)	Grass 1 (collect leachate)
S11	C11	L11	P11	Grass 2	Grass 2
S12	C12	L12	P12	Grass Control	Grass Control
S13	C13	L13	P13	Strawberry 1	Strawberry 1
S14	C14	L14	P14	Strawberry 2	Strawberry 2
S15	C15	L15	P15	Strawberry Control	Strawberry Control
S16	C16	L16	P16	Potatoes 1	Cabbage 1
S17	C17	L17	P17	Potatoes 2	Cabbage 2
S18	C18	L18	P18	Potatoes Control	Cabbage Control
Lysimeters					
Sa		La	Pa	Potatoes	Carrot, Cabbage and Dwarf French Beans
Sb		Lb	Pb	Carrots and Grass	Potatoes and Grass

Table 2 Varieties used

Crop	Variety	Crop	Variety
Dwarf French Beans	The Prince	Grass	Twystar Lolium
Cabbage	Golden Acre	Strawberry	Korona
Carrot	Early Nantes 5	Potatoes	Wilja

2.3 Supporting analyses

The determination of tritium in solid environmental materials was based on controlled combustion followed by liquid scintillation counting using a low-level Quantalus instrument that had been suitably calibrated. The analytical procedure had previously been tested via participation in an intercomparison exercise organised by the National Physical Laboratory. The results from HPA-RPD were in good agreement with the expected values.

The analytical reproducibility was demonstrated by replicate measurements made on one sample of potato from each lysimeter, grown as part of the 2005 study. Activity concentrations in potato flesh should be uniform. The results are shown in Table 3. In

all cases there was no significant difference between replicate measurements, which strongly supports the reproducibility of the analytical procedure.

Table 3 Total tritium concentration in potatoes grown in lysimeters, Bq g⁻¹

	Loam soil	Peat soil	Sandy soil
Measurement 1	0.143 ± .0020	0.080 ± 0.014	0.225 ± 0.026
Measurement 2	0.148 ± 0.021	0.102 ± 0.017	0.226 ± 0.030
Measurement 3	0.135 ± 0.021		

The detailed analytical procedure was as follows. All crop and soil samples were analysed via controlled combustion in a Raddec® pyrolyser. First, the temperature was raised slowly to 180°C and maintained at that level for 45 minutes. This step removed water from the sample. A stream of air carried the water vapour over a catalyst held at a temperature of 500°C and then into a trap containing a known volume of dilute nitric acid solution. At the end of this cycle the solution in the trap was replaced with fresh dilute nitric acid. The temperature of the sample itself was then raised to 500°C. The air was then replaced by oxygen and the sample maintained at 500°C in the flow of oxygen for at least an hour.

The solution in the trap after the lower temperature phase was defined as containing the aqueous tritium from the sample; that from the higher temperature phase was defined as the organically bound tritium (OBT). These are operational definitions, ie they relate to the conditions under which the procedure is carried out.

For the solutions from each trap, an aliquot was taken and mixed with scintillant and counted in the Quantulus against a reagent blank prepared using the same dilute nitric acid solution that was placed in the traps. The pyrolyser efficiency was checked regularly by analysing an aliquot of standardised tritiated thymidine solution; the efficiency of counting was determined using a standardised aqueous tritium standard.

In all cases the aliquots used for the analyses were about 10 g for crops and about 5 g for soil. All results were calculated as Bq g⁻¹ with respect to fresh mass for crops and “as collected” mass for soils. The concentrations of tritium in samples for 2006 were expected to be lower than those in 2005. For this reason, samples from 2006 were analysed using a slightly modified method in which a smaller volume of dilute acid was used and the trap was cooled. The aim of this modification was to improve the limit of detection.

Samples of rainwater and leachate were prepared for counting using distillation. Where total tritium was required, potassium permanganate was added and the sample boiled under a reflux condenser for 30 minutes before distilling an aliquot for measurement. The distillates were prepared and counted in the same way as the solutions in the traps from the pyrolyser. In this case however distilled water was used as a reagent blank.

In this report, all individual results have been reported together with their uncertainties. The reported uncertainties were based on standard analytical uncertainties multiplied by a coverage factor of k=2. This provides a level of confidence of approximately a 95% confidence interval. However, no allowance has been made for the uncertainty arising from sampling.

3 RESULTS AND DISCUSSION

All crops were sown or planted as detailed in Tables 1 and 2. Unfortunately, in 2005 the only strawberry plants that produced any fruit were those in the control containers, and so no results were available for this crop. All other crops produced sufficient material for analysis. The growing season in 2006 was exceptionally hot and dry and therefore all crops required extensive watering with tap water. Despite this, very little leachate was collected from the containers set up for this purpose. In addition, yields of all crops were lower than in 2005. In particular the hearts of the cabbages were poorly formed and one of the carrot crops grown on clay failed completely.

3.1 Concentrations of tritium in soil

The results for total and aqueous tritium concentrations in soils from the lysimeters are shown in Figures 1 – 3. These data relate to the surface layer of the soil, ie the depth over which the pellets had originally been incorporated. The Figures include all of the data from the 2005 and 2006 growing seasons, together with those from single samples of each soil type taken in June 2007. The detailed data for individual lysimeters and small containers are given in Tables A1 and A2.

In Figures 1 – 3 the total tritium data are given in blue and the corresponding values for aqueous tritium are shown in red. Both sets of data indicate a large variability in the individual results. However, this variability tended to decrease with time after the pellets were added. The reproducibility of the analytical method has been established (Section 2.3 and Table 3). Consequently, the variability can probably be ascribed to the non-uniform distribution of sludge in the soil. In the early stages following the amendment of the soil, individual sludge pellets were visible in the sample and some passed through the sieve intact. This would obviously lead to problems with sample homogeneity. The gradual break up of pellets is consistent with the reduced variability over time.

The soils used for growing grass were the only ones that were not disturbed by cropping or tilling between the two years. For this reason, these results are differentiated in Figures 1 – 3 by using square points. However the data for these soils did not appear to be less variable than the other results.

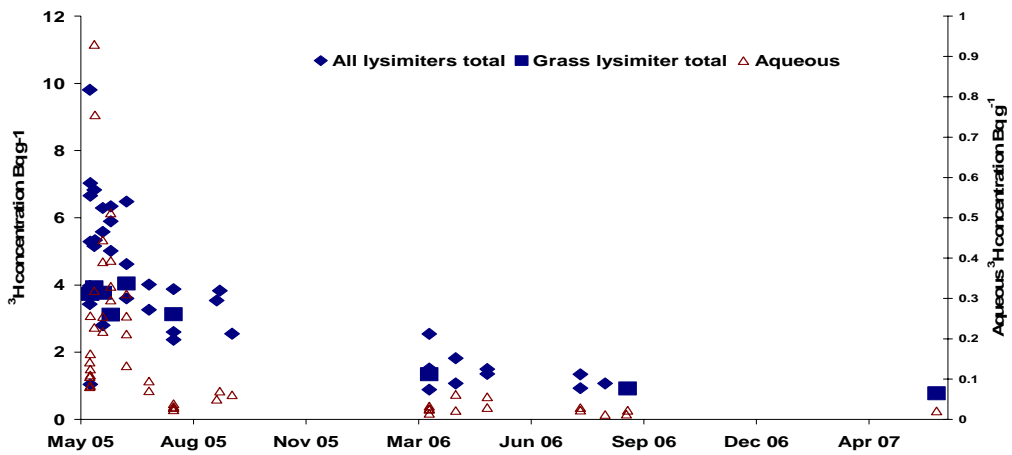


Figure 1 Lysimeter: concentration of tritium in peat soil

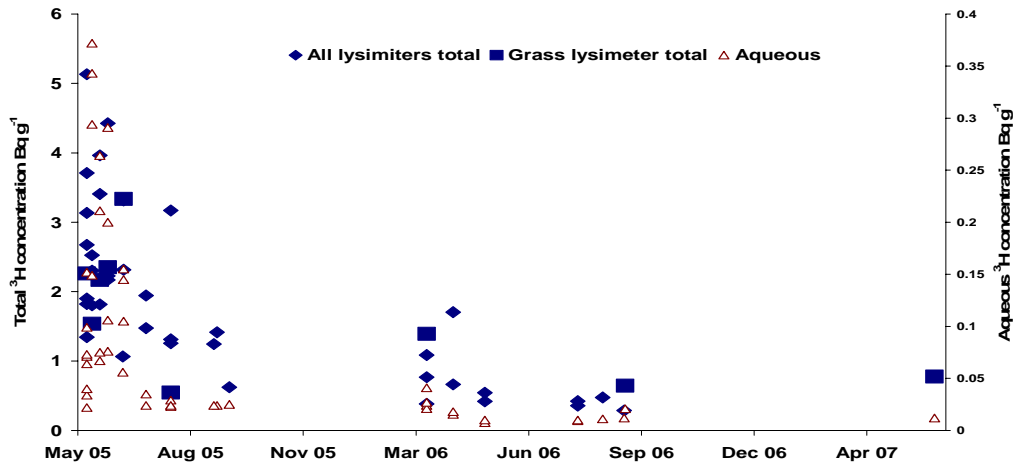


Figure 2 Lysimeter: concentration of tritium in loam soil

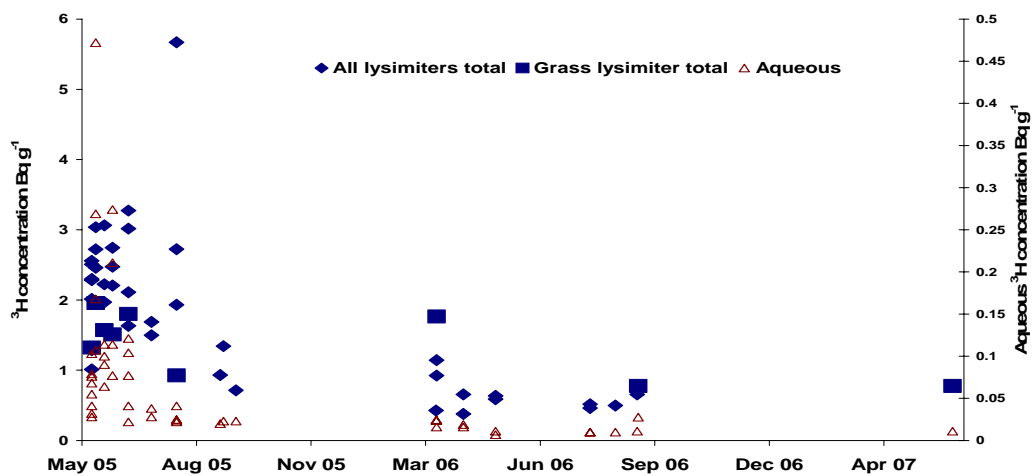


Figure 3 Lysimeter: concentration of tritium in sandy soil

Figures 4 – 7 show the results for total tritium in the four soil types studied in the small containers. Sampling and measurements were confined to the treated soils. Although these soils were only sampled at the beginning and end of the growing seasons, the same pattern of both decreasing variability and concentration with time was evident.

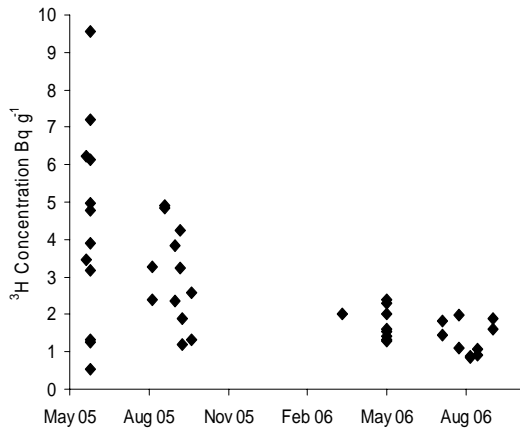


Figure 4 Containers: concentration of tritium in peat soil

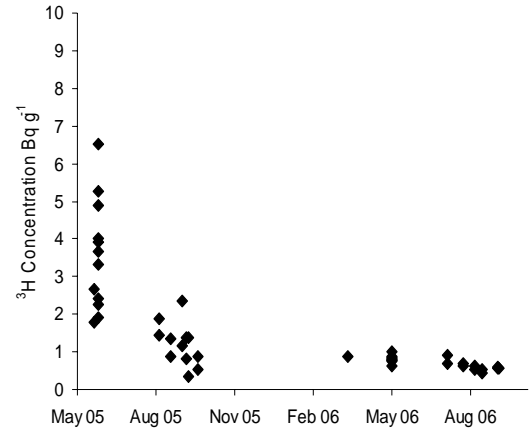


Figure 5 Containers: concentration of tritium in loam soil

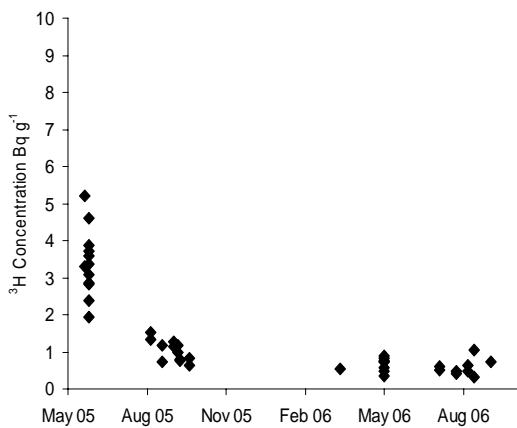


Figure 6 Containers: concentration of tritium in sandy soil

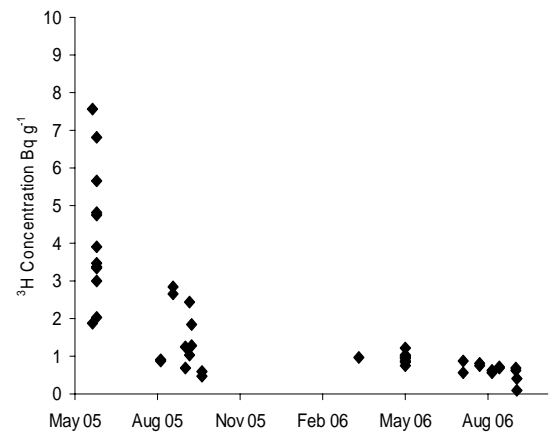


Figure 7 Containers: concentration of tritium in clay soil

The non-uniform distribution of the sludge pellets in the soil in the early stages of the study mean that only very broad comments can be made about temporal trends. The inhomogeneity decreased with time but the ranges of individual values from the same soil were still large. This explains the apparent increase in the mean tritium concentration in sandy soil during 2006 seen in Table 4. The ranges of the two data sets (April/May and August/September) overlapped considerably and so the means were not significantly different from each other.

Figures 1 – 3 indicate that the concentrations of aqueous tritium were much lower than the corresponding values for OBT. There was considerable variability in the values for aqueous tritium, but nevertheless the indications were that the concentration of aqueous tritium decreased much more rapidly over time than the corresponding value for total

tritium. Within 3 months of the initial application of sludge pellets the measured concentrations of aqueous tritium in all soils were very close to the limit of detection and remained at this level throughout the remainder of the project.

Table 4 Change in activity concentrations of tritium in soils over time

Soil type and Period	Average concentration for the period, Bq g ⁻¹			% decrease in the growing season	
	Aqueous	Organically bound	Total		
Lysimeters					
Loam	May / June 2005	0.14	2.47	2.61	
	September 2005	<0.02	1.07	1.09	58%
	April / May 2006	0.01	0.82	0.83	
	August / September 2006	0.01	0.45	0.46	44% (82% in both years)
	June 2007 (single value)	0.01	0.63	0.64	
Peat	May / June 2005	0.29	4.70	4.99	
	September 2005	0.06	3.25	3.31	34%
	April / May 2006	0.04	1.40	1.44	
	August / September 2006	0.02	0.95	0.99	33% (81% in both years)
	June 2007 (single value)	0.02	0.87	0.89	
Sand	May / June 2005	0.12	2.09	2.21	
	September 2005	<0.02	0.97	1.00	55%
	April / May 2006	<0.01	0.55	0.56	
	August / September 2006	<0.02	0.63	0.67	-17% ¹ (71% in both years)
	June 2007 (single value)	0.01	0.95	0.96	
Containers					
Loam	May 2005	0.27	3.28	3.55	
	August / September 2005	0.02	1.21	1.23	65%
	May 2006	0.01	0.81	0.82	
	August / September 2006	0.01	0.54	0.55	32% (84% in both years)
Peat	May 2005	0.34	4.04	4.38	
	August / September 2005	0.05	3.17	3.22	27%
	May 2006	0.06	1.88	1.94	
	August / September 2006	0.03	1.26	1.29	34% (71% in both years)
Sand	May 2005	0.16	3.23	3.39	
	August / September 2005	0.02	1.11	1.13	67%
	May 2006	0.01	0.65	0.66	
	August / September 2006	0.01	0.58	0.59	11% ¹ (83% in both years)
Clay	May	0.36	3.86	4.22	
	August / September 2005	0.02	1.38	1.40	67%
	May 2006	0.03	0.92	0.95	
	August / September 2006	0.02	0.53	0.55	42% (87% in both years)

Note 1 Given the spread of the individual data for the sand soil in May and August/September 2006 (Appendix A), the mean values given here are not significantly different from each other.

Table 4 gives a comparison of the average concentrations in surface soil over time for both the lysimeters and the small containers. The values for May and June 2005 include all of the samples collected during the first few weeks after the soils had been

amended. For the lysimeters, the data for single samples taken in June 2007 are also included. In very broad terms, the average concentrations of total tritium in surface soils decreased with time. By the end of the second growing season, on average the sandy soil in lysimeters had lost about 70% of total tritium initially added at the start of the experiment. The corresponding changes in the peat and loam soils were both about 80%. Similar changes in average values were observed for soils in the small containers, with the concentration in clay soil having lost about 85% of the added tritium.

3.2 Transport of tritium in the soil

3.2.1 Migration down the soil column

The samples used to derive the data in Figures 1 – 3 were taken from the surface of the soil. The size of the lysimeters was sufficient for soil cores to be taken just before the harvesting of crops in August 2005 and again in April 2006. Concentrations of tritiated water and OBT were determined at different depths in the soil, and the results are summarised in Table 5.

Table 5 Variation in concentrations of tritium with depth in lysimeters

Depth, mm	Measured Tritium, Bq.g ⁻¹					
	Loam		Peat		Sandy	
	Aqueous	Organically Bound	Aqueous	Organically Bound	Aqueous	Organically Bound
August 2005						
0 – 100	0.04 ± 0.02	0.67 ± 0.07	0.03 ± 0.02	1.2 ± 0.1	<0.02	0.70 ± 0.08
100 – 200	0.02 ± 0.01	0.10 ± 0.02	<0.02	0.35 ± 0.04	<0.02	0.04 ± 0.01
200 – 300	0.03 ± 0.02	0.02 ± 0.01	0.03 ± 0.02	0.04 ± 0.02	<0.02	<0.02
April 2006						
0 – 100	<0.02	0.38 ± 0.05	0.04 ± 0.02	0.9 ± 0.1	<0.02	0.8 ± 0.1
100 – 200	<0.02	0.04 ± 0.01	0.05 ± 0.01	0.24 ± 0.03	<0.02	0.03 ± 0.01
200 – 300	<0.02	0.05 ± 0.01	0.06 ± 0.02	0.15 ± 0.03	<0.02	<0.02

The results for 2005 indicated that, for all of the three soil types studies, most of the activity retained in the soil was still in the surface layer (0 – 100 mm) and was mostly in the form of OBT. The results for 2006 also indicated that generally most of the activity in the soil was in the form of OBT, and again most of the activity was still in the surface layer. The one exception was the peat soil, where there was some evidence of migration of OBT to 300 mm depth.

Overall, most of the activity in the soils was in the form of OBT, and this mostly remained in the surface layer. These results indicated therefore that the overall decreases in activity concentrations in surface soil (Section 3.1) were not due to the migration of OBT down the soil column.

3.2.2 Transport out of the soil in leachate

As noted in Section 2.2, the small containers provided an opportunity to collect leachate, ie the fluid that had percolated through the soil. For each soil type, leachate was

collected from one of the small containers that had been treated with sewage sludge pellets and then used to grow grass. These were considered the most suitable because after the crop had been sown the underlying soil would not be disturbed again throughout the duration of the study.

The leachate being produced was monitored regularly over the growing seasons in 2005 and 2006. No leachate was collected over the winter of 2005/6. Each time that leachate was collected the volume produced was measured so that a total flux of tritium out of the container could be estimated. In most cases analyses were confined to total tritium as described in Section 2. For a few samples, a separate aliquot was distilled without potassium permanganate to determine the tritiated water content. For these samples, there was no significant difference between the concentrations of aqueous and total tritium. It would be reasonable to assume therefore that most or all of the tritium lost via downward leaching was in the form of tritiated water.

The concentrations of tritium in leachate and the total lost from the containers over the 2005 growing season are detailed in Table A3. In 2006 the very hot and dry weather meant that very little leachate was produced. Consequently, it was only possible to obtain between one and three samples from each container throughout the season. These results are also included in Table A3. All data are given in terms of total tritium. In each case the volumes of leachate produced and the total activity lost are also given. Results for the two years are summarised in Table 6.

Table 6 Tritium in leachate

Soil type	Season	Volume, l	Tritium content, Bq	% of tritium added to container
Clay	2005	80.5	5,100 ± 400	23%
	2006	7.1	220 ± 20	1%
Loam	2005	70.7	3,300 ± 300	25%
	2006	2.8	130 ± 20	1%
Peat	2005	65.0	1900 ± 200	9%
	2006	11.5	240 ± 30	1%
Sandy	2005	61.3	4,600 ± 400	21%
	2006	1.9	66 ± 9	1%

All soil types showed a similar pattern of tritium loss in leachate with time. After a short period of about a month the concentration of tritium in the leachate peaked at about 0.1 – 0.2 Bq ml⁻¹. It then fell over the next month or so to a low and fairly constant value of about 0.01 – 0.02 Bq ml⁻¹.

Over the 2005 growing season, the total amount of tritium in the leachate from the peat soil was about 10% of the amount initially applied to the soil (Table 6). From Table 4, over the same period about 30% of the total tritium activity was lost from the surface layer of the peat soil. For each of the other soil types, about 20% of the activity originally applied was found in the leachate over the 2005 growing season, whereas the loss of activity from the surface layer over the same period was in the range 55 – 70% (Table 4). The changes in the amounts remaining in the surface soils were of necessity very broad estimates (Section 3.1). However, it seems very likely that the decreases in

activity from the surface soils cannot be accounted for solely by the activity transported out of the soil in leachate.

The data in Table 5 indicate that most of the activity remaining in the soil was retained in the surface layer in the form of OBT. This seems reasonably consistent with the fact that the fluxes of tritiated water out of the small containers were lowest in the late summer 2005 at about the time that the first set of soil cores were taken from the lysimeters (Table 5).

The extremely dry and hot weather in the summer of 2006 meant that only small volumes of leachate were collected. Over the growing season of 2006 only about 1% of the initially applied activity was transported out of the soil as leachate (Table 6).

3.2.3 Potential importance of deposition in rainfall

Rainfall was monitored on a regular basis to investigate whether activity concentrations observed in this study contained significant contributions from ambient levels of tritium in the general environment. The concentration of tritium in rainwater was measured throughout the growing seasons, and the results are given in detail in Table A4. One sample collected in August 2005 gave an anomalous measurement of 0.03 ± 0.04 Bq ml⁻¹. A similar though not a marked increase was seen in August 2006. All other measurements were less than or very close to the detection limit, which was 0.006 Bq ml⁻¹. The periods when the high measurements were observed coincided with the harvesting of both the carrots and potatoes. The rainfall collector was situated only a few metres away from the lysimeters, and there is a possibility that a small amount of soil may have been blown into the collector. Overall however, the measurements indicate that any tritium in rainwater was not contributing significantly to the concentrations observed in the leachate, nor to the surface soil. The concentrations of tritium in rainwater were very low, and for this reason no attempt was made to analyse the aqueous and organically bound fractions separately.

3.3 Transfer of tritium into vegetables

Tables A5 – A9 give the measured concentrations of tritium in vegetables, expressed in terms of fresh mass. The measured concentrations for crops grown in untreated soils were generally very low, many being below the limit of detection. These results imply that any cross-contamination via transpiration from treated soils and crops was small. They also suggest that any effects from the presence of ambient levels of tritium in the general environment are small.

All measured values for vegetables grown in treated soils were low, with large associated counting uncertainties. Generally, however, where analyses were carried out on the same crop and soil type in two separate containers, the results were not significantly different from each other. In most cases the tritium in vegetables was nearly all present as tritiated water. Where this was not the case, for example in dwarf French beans grown in loam soil in 2005, the same trend was not observed in the same crop when grown in other soil types, nor in the same soil type at a different time. The greater proportion of OBT in certain samples could be the result of small amounts of soil

being attached to the vegetation, since most of the tritium in the surface soil was in the form of OBT throughout both of the growing seasons (Section 3.1). The concentration of OBT observed in the beans grown in loam soil in 2005 does however seem too great to be explained by soil contamination, because from Table 4 and Tables A1 and A2 it would imply that around 5 – 10% by mass of the sample analysed was soil. This seems unlikely given that all vegetables were washed prior to analysis. This result must be regarded as anomalous given that the tritium content of another sample of beans collected on the same day from an adjoining small container was mainly in the form of tritiated water.

The absence of OBT in many samples of vegetables deserves further comment, because tritium should become incorporated into organic matter as a result of photosynthesis. The moisture content of most crops is between 80 and 95%. On the basis of the specific activity approach the $^1\text{H} : ^3\text{H}$ ratio in the plant water and organic matter should be the same (IAEA, 2001). Taking the relatively low percentage of organic matter in fresh vegetables with the very low concentrations of tritiated water in some of these samples, then concentrations of tritium in the form of OBT might be below the detection limit.

The potential importance of the distribution of activity between the peel and flesh of potato tubers was noted in Section 2.2. Data for several other radionuclides have already been published and the implications for radiological assessments have been discussed (Green et al, 1997; Ham et al, 1998). The concentrations of tritiated water and OBT in the peel and flesh of one sample of potatoes are given in Table A7. Although the data for OBT might imply some contribution from soil, the tubers were washed thoroughly prior to peeling. The data in Table A7 indicate that concentrations of tritiated water in the peel and flesh of potato tubers were not significantly different. The corresponding values for OBT were also not significantly different, although the uncertainties in these data were considerable. Overall, these limited data suggest that in radiological assessments for tritium there is no need to take account of whether potatoes are consumed after peeling or as whole tubers.

In order to aid comparisons Table 7 gives the average concentrations of tritium (as tritiated water) in crops for each container - soil - crop combination. For the small containers, the variability in concentrations of tritium between the same crop grown in different soil types were fairly small and were comparable with the variability between replicate experiments involving the same soil and the same crop (Tables A5 – A9).

In 2005, only potatoes and carrots were grown in both the lysimeters and in the small containers. For both crops the activity concentrations in crops grown in lysimeters were generally significantly greater than the corresponding values for those grown in the small containers. In 2006, all crops except strawberries were grown in both the lysimeters and the smaller containers. Nearly all the measured concentrations were lower than those for 2005 and in many cases were very close to those measured in the control crops. In addition, the large difference in the concentrations of tritium in potatoes and carrots grown in lysimeters and containers observed in 2005 was not apparent in the crops grown in 2006.

For many radionuclides, uptake from soil into plants has been defined using the soil – plant transfer factor approach, ie the quotient of the activity concentration in the plant to that in the soil. In the case of tritium, uptake by plants is likely to occur via chemical forms such as tritiated water that would be in the soil solution. The decreases in concentrations in vegetables between 2005 and 2006 could be linked with the decrease in the concentration of tritiated water in the surface soil over time (Figures 1 – 3). However, concentrations of tritiated water and OBT in surface soil changed rapidly during the 2005 growing season, and any quantitative assessment would require the use of a dynamic predictive model. Such work is outside the remit of the present study.

In qualitative terms, it would be reasonable to expect that tritiated water would be produced as part of the decomposition of the OBT. The rate of production would be dependent on factors such as temperature and the actual chemical forms of the OBT. In addition, the factors affecting the rate of removal of tritiated water from the surface soil would include rates of rainfall and any artificial irrigation, temperature and cover by foliage. Taking these factors together, the conventional soil – plant transfer factor approach would not be appropriate for tritium applied to land as sewage sludge.

The aim of this project was to investigate the movement of tritium into the edible parts of plants. The total amount of activity transferred into the vegetation, including the root systems, has not been studied specifically. However, the total amount of vegetation produced in each of the individual experiments in the small containers would be no more than a few kg fresh mass. Taking a broad and cautious value of 10 kg fresh mass of vegetation together with the concentrations in vegetation for 2005 (from Table 7), the total amount of tritium removed from the soil via transfer into a vegetable crop would be of the order of 500 Bq. Taking this very broad estimate together with the amount taken out of the soil via leachate over the 2005 growing season (Section 3.2) and the amount remaining in the soil (Section 3.1) still does not account for all of the activity that was added. The likelihood is that tritium will have been lost via transpiration into the atmosphere, either from the soil, or from the vegetation during growth, or via a combination of the two. A rigorous investigation of this process would require dedicated and expensive equipment and was outside the scope of this project.

To provide some context, an aggregated transfer quotient approach might be helpful to provide an indication of the overall transfer of tritium to crops from the activity originally applied in this study. For this purpose, a quotient has been estimated that relates the concentration in the edible part of the crop in Bq kg^{-1} fresh mass to the original amount of activity applied to the soil in Bq m^{-2} . This quotient would then take account of the processes by which tritium was lost from the system for these particular experimental conditions such as rainfall rates and temperature. Consequently, the numerical values of such quotients would only be appropriate for specific circumstances and should not be applied more generally in radiological assessments. However, this approach does illustrate the level of transfer observed in this study. Table 8 lists the values of this aggregated transfer quotient (ATQ) that have been derived for the crops grown in 2006 in the lysimeters, ie for crops that were grown in broad conformance with the requirements of the Safe Sludge Matrix. The aggregated transfer quotients were broadly similar between crop and soil types. The observed values were typically around $2 \cdot 10^{-4} \text{ m}^2 \text{ kg}^{-1}$, which illustrates how little of the activity originally applied was transferred to crops. On the basis of these indicative values, for general assessments it would be

reasonable to assume that only a small proportion of the tritium in soil treated with sewage sludge would be transferred into crops.

Table 7 Average concentration of tritiated water in crops

Soil	Experiment	Tritium concentration, Bq g ⁻¹					
			Cabbage	Carrots	DF Beans	Potatoes	Strawberries
Clay	2005	Container	0.035	0.025	0.04	0.04	
	2006	Container	<0.007	0.03	0.023	0.010	0.014
Loam	2005	Container	0.05	0.016	0.05	0.035	
		Lysimeter		0.10		0.12	
	2006	Container	0.01	0.03	0.029	0.016	0.015
		Lysimeter	0.033	0.04	0.05	0.024	
Peat	2005	Container	0.025	0.03	0.05	0.05	
		Lysimeter		0.12		0.08	
	2006	Container	<0.008	0.02	0.023	0.027	0.017
		Lysimeter	0.032	0.03	0.039	0.057	
Sandy	2005	Container	0.05	0.05	0.05	0.07	
		Lysimeter		0.11		0.20	
	2006	Container	0.01	0.03	0.033	0.033	0.015
		Lysimeter	0.048	0.02	0.037	0.040	
Controls (all types)	2005	Container	<0.02	<0.02	0.05	<0.02	
	2006	Container	<0.01	0.018	0.02	0.01	0.012

Table 8 Aggregated transfer quotient for 2006 lysimeter crops

Soil	Quotient x 10 ⁴ (Bq kg ⁻¹ in fresh crop / Bq m ⁻² applied to soil)			
	Cabbage	Carrots	DF Beans	Potatoes
	Loam	1.9	2.4	2.9
Peat	1.9	1.8	2.2	3.4
Sandy	2.8	1.2	2.2	2.4

An aggregated transfer approach has also been adopted in a lysimeter study where tritiated water was applied to soil at different stages of growth of a crop of Chinese cabbage (Choi et al, 2007). The entire experiment was completed within about 80 days. The values for the ATQ were low, in the range $1 \times 10^{-5} \text{ m}^2 \text{ kg}^{-1}$ to $5 \times 10^{-3} \text{ m}^2 \text{ kg}^{-1}$ in terms of the HTO in fresh plant tissue. Generally, the observed values decreased as the time between the application of the tritiated water and the harvest increased. There was some transport of activity down into the soil but most of the activity originally applied was lost relatively quickly. For example, of the activity originally applied to the soil 26 days after sowing, only 0.5% remained in the soil 8 weeks later. Estimates of the total activity in the plants and in the soil at the end of the experiment indicated that most of the applied HTO had been lost via transpiration into the atmosphere before the plants were ready for harvest. A simple comparison of ATQ values from this published work

and from the study described in this report indicates that in both cases the transfer of tritium from soil into crops was small. However, the factors that contributed to the limited transfer in each case deserve examination. In the published study dealing with a surface application of HTO, transpiration was the dominant process with losses occurring over a period of a few weeks. In the present project, where tritium-bearing sludge was incorporated into the soil, transpiration was still important but so was the short term leaching of activity out of the soil as HTO and the longer term retention of OBT in the surface soil. On this basis, the form in which the tritium has been applied to the soil needs to be taken carefully into account when using published information to develop or validate predictive models.

Appendix B gives a brief comparison of the ATQ derived from the work on sewage sludge with values that can be distilled from a predictive model used by RPD. In the short term after the activity had been applied to the soil, the model predicted a more rapid transfer than was observed in this study, although the average ATQ over about a 2-year period was less than twice the observed value. In contrast, in the longer term the predicted ATQ was very much less than the observed value. These differences were attributed to the assumption in the model that the tritium had been applied to the soil in the form of HTO and HT, rather than as OBT. In the short term, the model would therefore predict that all of the activity would be available for uptake. However, the model would also predict that the activity would be lost from the soil relatively quickly, and so ATQ values in the longer term were very low. In practice, where the tritium has been applied to the soil in the form of OBT, much of the activity remains in that form in the surface soil (Section 3.2.1).

3.4 Grass

All of the results for grass are given in Table A10. In contrast to the vegetable crops where most of the activity was in the form of tritiated water, the results for grass indicated a significant and variable contribution from OBT. As in the case of arable crops, grass has a high moisture content and so the amount of OBT deriving solely via root uptake from the soil is likely to be close to or below detection limits (Section 3.3). The amounts of OBT observed in grass therefore suggest that a considerable proportion of the measured tritium activity was due to soil being splashed on to the vegetation. This process has been evaluated in studies of other radionuclides (Green et al, 1996). In addition, it was identified as potentially important in the recent review on the use of sludge on agricultural land carried out by the NRPB on behalf of FSA (Ham et al, 2003). The effect might be expected to be more marked in this study because the grass had only been sown shortly before sampling began and the sward had not yet developed fully.

Taking broad averages, the OBT content of the grass could be accounted for by the presence of a few percent by mass of the underlying soil. However, because the aqueous tritium : OBT ratio in surface soil is very low, a few percent of soil in the grass by mass would not significantly affect the measured concentration of aqueous tritium. This is best illustrated by a worked example. Taking the loam soil in the lysimeter in 2005, the tritium in grass was measured on 27/6/05 as aqueous = 0.05 ± 0.02 and OBT

= $0.07 \pm 0.01 \text{ Bq g}^{-1}$ (Table A9). Soil was sampled from this lysimeter on 17/6/05. The analysis gave a concentration of aqueous tritium of $0.16 \pm 0.02 \text{ Bq g}^{-1}$ and a value for OBT of $3.2 \pm 0.3 \text{ Bq g}^{-1}$ (Table A1). On this basis, about 2% of soil by mass in the grass would account for all the observed OBT. This is around the percentage that has been observed in field studies on established pasture (Green et al, 1996). It should be noted however that the same percentage of soil by mass would account for only about 3% of the aqueous tritium in the grass.

In practical terms, the effect of soil splash also appeared to mask any differences between grass grown in the lysimeters and that grown in treated soils in the smaller containers. In addition, while most other crops grown in untreated soils gave measured values close to or below the limit of detection, measurable values were obtained for some of the blank samples of grass, in some cases for both aqueous tritium and OBT. Again, concentrations of OBT were measurable which suggests that some contamination with treated soil had occurred. The analyses of the control samples were repeated and the results were in good agreement with the earlier values. All of the measured concentrations of tritium in soils from the untreated (control) containers were either at or close to the limit of detection (Table A2), which indicated that the untreated soils themselves had not become contaminated. Precautions had been taken against cross contamination both while growing the grass and during collection of the samples, but these were clearly insufficient.

The results for grass, together with the retention of OBT in the surface soil, have implications for predictions of the onward transfer of activity into grazing animals. For many radionuclides, the inadvertent ingestion of soil during grazing can be an important route of intake of activity (see for example Green et al, 1996). The results from this study indicate that for tritium, soil ingestion could be important when new pasture is sown on land that has been treated with sludge. This is because uptake of tritium as tritiated water from the soil is relatively low while a reservoir of activity remains in the surface soil in the general form of OBT. The availability of tritium in an organically-bound form for onward transfer to milk or meat would be difficult to quantify since it could depend on the actual chemical species involved.

Sewage sludge can also be applied to existing pasture, either directly on to the herbage in the case of enhanced treated sludge or via direct injection for conventionally treated material. In both cases, grazing cannot take place until at least 3 weeks after the sludge has been applied. Published information has been reviewed, and the conclusions were that it would be prudent for predictive models to consider the ingestion of sludge-associated radionuclides even after direct injection into the soil (Ham et al, 2003). On the basis of the present study, inadvertent ingestion of sludge could be important for tritium.

4 CONCLUSIONS

Concentrations of tritium in treated soil decreased throughout the period of the experiment. Measured values in the soil itself displayed considerable variability,

although this decreased with time. The trend in improved homogeneity with time might have been expected given the form of the activity applied and the relatively short time between the application of the sludge pellets and the initial measurements.

The variability between samples made the study of temporal changes difficult. In broad terms however about 80% of the tritium originally applied was lost over the two growing seasons, with the rate of loss appearing to be lower in the second season. The tritium that remained in the soil was largely in the form of OBT. Studies of the distribution with depth showed that generally the activity was retained in the surface (0 – 100 mm) layer. The concentrations of aqueous tritium in soil climbed rapidly to a peak and then fell to a near constant level within about 2 to 3 months of application.

The results of measurements in leachate from the small containers indicate a marked loss of tritium in the form of tritiated water particularly during 2005. Much less tritium was lost in 2006, but this may have been due to the extreme drought. However, in both years, the total amount of activity measured in leachate was insufficient to account for losses implied from the measurements in surface soil. In particular, during 2006 very little tritium was measured in leachate, possibly because of the drought, yet all the containers showed a significant loss of tritium soil activity. When taken with the retention of OBT in the surface layer of the soil and the amount of activity that was estimated to be in the crops as a whole, it seems likely that activity was lost via transpiration from the soil or from the crop, or from a combination of the two. A much more sophisticated experiment would be needed to quantify losses via transpiration, and this was outside the scope of the present study.

Concentrations of tritium were measurable in nearly all crops grown in treated soils, but the actual values were generally low. With the exception of grass, the activity was mainly in the form of tritiated water. However, all of the crops studied had a high water content, and if the $^1\text{H} : ^3\text{H}$ ratio was similar in the aqueous and organic fractions of the vegetation, then in many cases the organically bound tritium could be below the limit of detection of the analytical method.

Samples of grass generally contained higher amounts of OBT than the vegetable crops. The concentrations of OBT were variable. Overall, the indications were that soil splash was an important contributor to the activity observed in the vegetation. Inadvertent ingestion of soil could therefore be an important contributor to intakes of tritium by animals grazing on pasture that had been treated with sludge. Predictions of onward transfer to milk or meat would then require information on the availability of tritium in an organically-bound form. This would be difficult to quantify since it could depend on the actual chemical species involved.

With the exception of grass, activity concentrations in all crops grown in untreated soils were near to, or below the limit of detection. These results indicated that contamination via transpiration from nearby treated crops or soil was not significant. Measurements in rainfall indicated that ambient levels of tritium in the environment were not sufficient to affect these experiments. The results for grass grown in the control containers indicated that some contamination with soil had occurred. The reasons for this were not clear.

For crops grown in treated soils in small containers, the reproducibility in the measured concentrations between individual soil - crop combinations was good. There were no

significant differences in the concentrations of tritium in the same crop grown in different soils. Differences between crops grown at the same time in the same size of container were also small. However, in 2005 the concentrations of tritium in crops grown in lysimeters were significantly greater than the corresponding values in the small containers, by factors of between 2 and 5. This effect was not as evident in 2006 but in general the crops grown in the lysimeters did contain more tritium than those grown in containers. The larger physical size of the lysimeters means that they are likely to reproduce conditions in the field more closely than the small containers. In addition, in 2006 it was possible to grow enough of all the crops needed for this study in the lysimeters. However, the small containers provided an opportunity to expand the range of soil types being studied and to evaluate the loss of tritium via leachate. The results from the small containers therefore provided valuable support to the work done with the lysimeters.

The changes in concentrations of tritium in soil over the growing seasons and the low concentrations measured in crops mean that it was not possible to quantify individual soil to crop transfer in terms of the conventional soil – plant transfer approach that has been applied to many other radionuclides. Any quantitative assessment of the experimental data would require the use of a dynamic predictive model. Such work was outside the remit of the present study. However, an aggregated transfer quotient approach has been used to relate the concentration in the fresh edible parts of crops grown in 2006 and the amount of activity originally applied to the soil in 2005. The data for crops grown in 2006 were used because of the constraints of the Safe Sludge Matrix. The aggregated transfer quotients were broadly similar between crop and soil types, the observed values being typically around $2 \times 10^{-4} \text{ m}^2 \text{ kg}^{-1}$. The derived parameter values relate specifically to the conditions encountered in the present experiment and should not be used more generally. However, on the basis of these indicative values, for broad assessment purposes it would be reasonable to assume that the proportion of tritium transferred into crops from soil treated with sewage sludge would be small.

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APPENDIX A Analytical results

A1 TRITIUM IN SOIL

Table A1 Measured tritium concentration in lysimeter soils

Soil		Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)		
				Aqueous	OBT	Total
Loam	2005	La (16/5)	13%	0.03 ± 0.01	2.64 ± 0.28	2.67 ± 0.28
		La (16/5)	17%	0.07 ± 0.02	1.75 ± 0.19	1.82 ± 0.19
		La (16/5)	17%	0.15 ± 0.02	4.98 ± 0.37	5.13 ± 0.37
		La (16/5)	10%	0.10 ± 0.02	3.61 ± 0.28	3.71 ± 0.28
		La (20/5)	19%	0.29 ± 0.04	2.23 ± 0.23	2.52 ± 0.23
		La (20/5)	17%	0.34 ± 0.04	1.96 ± 0.21	2.30 ± 0.21
		La (27/5)	19%	0.07 ± 0.02	3.34 ± 0.35	3.41 ± 0.35
		La (27/5)	20%	0.26 ± 0.03	3.70 ± 0.39	3.96 ± 0.39
		La (3/6)	15%	0.08 ± 0.02	4.35 ± 0.45	4.43 ± 0.45
		La (3/6)	18%	0.20 ± 0.03	1.97 ± 0.21	2.17 ± 0.21
		La (17/6)	18%	0.06 ± 0.02	1.01 ± 0.11	1.07 ± 0.11
		La (17/6)	20%	0.15 ± 0.02	3.17 ± 0.33	3.32 ± 0.33
		La (7/7)	22%	<0.02	1.92 ± 0.24	1.94 ± 0.24
		La (29/7)	22%	<0.02	3.15 ± 0.06	3.17 ± 0.06
	La (29/7)	20%	<0.03	1.23 ± 0.04	1.26 ± 0.04	
	La (8/9)	18%	<0.02	1.39 ± 0.15	1.41 ± 0.15	
	La (13/3)	20%	<0.02	0.36 ± 0.05	0.38 ± 0.05	
	La (13/3)	18%	0.04 ± 0.02	0.73 ± 0.10	0.77 ± 0.10	
	La (6/4)	13%	0.02 ± 0.01	1.69 ± 0.03	1.71 ± 0.03	
	La (4/5)	9%	0.01 ± 0.01	0.54 ± 0.08	0.54 ± 0.08	
	La (25/7)	15%	<0.01	0.41 ± 0.06	0.42 ± 0.06	
	La (16/8)	20%	<0.01	0.46 ± 0.07	0.48 ± 0.07	
	La (4/9)	18%	<0.01	0.27 ± 0.04	0.29 ± 0.04	
	La (16/5)	10%	0.02 ± 0.01	2.24 ± 0.17	2.26 ± 0.17	
	La (16/5)	11%	0.04 ± 0.01	1.86 ± 0.20	1.90 ± 0.20	
	La (16/5)	12%	0.07 ± 0.02	3.06 ± 0.32	3.13 ± 0.32	
La (16/5)	9%	0.06 ± 0.01	1.28 ± 0.11	1.34 ± 0.11		
La (20/5)	19%	0.15 ± 0.02	1.39 ± 0.15	1.54 ± 0.15		
La (20/5)	17%	0.37 ± 0.05	1.43 ± 0.15	1.80 ± 0.16		
La (27/5)	14%	0.08 ± 0.02	1.74 ± 0.19	1.82 ± 0.19		
La (27/5)	16%	0.21 ± 0.03	1.96 ± 0.21	2.17 ± 0.21		
La (3/6)	14%	0.11 ± 0.02	2.12 ± 0.22	2.23 ± 0.22		
La (3/6)	16%	0.29 ± 0.04	2.06 ± 0.22	2.35 ± 0.22		
La (17/6)	17%	0.11 ± 0.02	2.21 ± 0.23	2.32 ± 0.23		
La (17/6)	18%	0.16 ± 0.02	3.18 ± 0.33	3.34 ± 0.33		
La (7/7)	17%	0.04 ± 0.02	1.44 ± 0.18	1.48 ± 0.18		
La (29/7)	19%	<0.02	1.29 ± 0.04	1.31 ± 0.04		
La (29/7)	17%	<0.02	0.52 ± 0.03	0.55 ± 0.03		

Table A1 Measured tritium concentration in lysimeter soils

Soil	Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)				
			Aqueous	OBT	Total		
Lb	2006	Lb (5/9)	13%	<0.02	1.22 ± 0.13	1.24 ± 0.13	
	Lb (19/9)	55%	<0.03	0.60 ± 0.08	0.62 ± 0.08		
	Lb (13/3)	20%	0.02 ± 0.02	1.37 ± 0.17	1.39 ± 0.17		
	Lb (13/3)	18%	<0.03	1.06 ± 0.14	1.09 ± 0.14		
	Lb (6/4)	13%	0.02 ± 0.01	0.65 ± 0.08	0.66 ± 0.08		
	Lb (4/5)	15%	<0.01	0.41 ± 0.06	0.42 ± 0.06		
	Lb (25/7)	14%	<0.01	0.35 ± 0.05	0.36 ± 0.05		
	Lb (5/9)	21%	0.02 ± 0.01	0.62 ± 0.09	0.64 ± 0.09		
	2007	Lb (7/6)	15%	0.01 ± 0.01	0.63 ± 0.09	0.78 ± 0.09	
	Pa	2005	Pa (16/5)	39%	0.08 ± 0.02	3.89 ± 0.41	3.97 ± 0.41
			Pa (16/5)	33%	0.11 ± 0.02	9.70 ± 1.00	9.81 ± 1.00
			Pa (16/5)	44%	0.14 ± 0.02	3.29 ± 0.26	3.43 ± 0.26
			Pa (16/5)	44%	0.11 ± 0.02	6.92 ± 0.05	7.03 ± 0.05
			Pa (20/5)	48%	0.32 ± 0.04	4.84 ± 0.50	5.16 ± 0.50
Pa (20/5)			50%	0.76 ± 0.09	4.58 ± 0.48	5.34 ± 0.49	
Pa (27/5)			49%	0.39 ± 0.05	5.90 ± 0.61	6.29 ± 0.61	
Pa (27/5)			47%	0.45 ± 0.05	2.36 ± 0.25	2.81 ± 0.26	
Pa (3/6)			38%	0.30 ± 0.04	4.72 ± 0.49	5.02 ± 0.49	
Pa (3/6)			35%	0.51 ± 0.06	5.83 ± 0.61	6.34 ± 0.61	
Pa (17/6)			44%	0.31 ± 0.04	6.17 ± 0.64	6.48 ± 0.64	
Pa (17/6)			42%	0.13 ± 0.02	4.49 ± 0.47	4.62 ± 0.47	
Pa (7/7)			45%	0.10 ± 0.02	3.92 ± 0.49	4.02 ± 0.49	
Pa (29/7)			44%	<0.02	2.35 ± 0.05	2.37 ± 0.05	
Pa (29/7)		44%	0.04 ± 0.02	3.84 ± 0.06	3.88 ± 0.06		
2006		Pa (8/9)	43%	0.07 ± 0.02	3.76 ± 0.39	3.83 ± 0.39	
		Pa (13/3)	45%	<0.03	0.86 ± 0.11	0.89 ± 0.11	
		Pa (13/3)	46%	0.03 ± 0.02	1.48 ± 0.19	1.51 ± 0.19	
		Pa (6/4)	39%	0.02 ± 0.01	1.80 ± 0.23	1.82 ± 0.23	
		Pa (4/5)	44%	0.03 ± 0.01	1.47 ± 0.20	1.50 ± 0.20	
		Pa (25/7)	26%	0.02 ± 0.01	1.32 ± 0.18	1.34 ± 0.18	
		Pa (16/8)	46%	0.01 ± 0.01	1.06 ± 0.15	1.07 ± 0.15	
		Pa (4/9)	41%	<0.01	0.89 ± 0.12	0.90 ± 0.12	
		2005	Pb (16/5)	43%	0.13 ± 0.02	0.92 ± 0.10	1.05 ± 0.10
			Pb (16/5)	29%	0.08 ± 0.02	3.65 ± 0.27	3.73 ± 0.27
			Pb (16/5)	34%	0.16 ± 0.03	5.13 ± 0.53	5.29 ± 0.53
			Pb (16/5)	36%	0.26 ± 0.04	6.40 ± 0.66	6.66 ± 0.66
			Pb (20/5)	48%	0.23 ± 0.03	3.70 ± 0.39	3.93 ± 0.39
	Pb (20/5)		47%	0.93 ± 0.10	5.90 ± 0.61	6.83 ± 0.62	
Pb (27/5)	45%		0.22 ± 0.03	3.55 ± 0.37	3.77 ± 0.37		
Pb (27/5)	42%		0.26 ± 0.03	5.33 ± 0.55	5.59 ± 0.55		
Pb (3/6)	36%		0.33 ± 0.04	2.79 ± 0.29	3.12 ± 0.29		
Pb (3/6)	41%		0.39 ± 0.05	5.50 ± 0.57	5.89 ± 0.57		
Pb (17/6)	42%		0.21 ± 0.03	3.39 ± 0.35	3.60 ± 0.35		
Pb (17/6)	43%		0.26 ± 0.03	3.79 ± 0.40	4.05 ± 0.40		
Pb (7/7)	47%		0.07 ± 0.02	3.19 ± 0.40	3.26 ± 0.40		
Pb (29/7)	49%		<0.03	2.57 ± 0.27	2.60 ± 0.27		

Table A1 Measured tritium concentration in lysimeter soils

Soil	Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)		
			Aqueous	OBT	Total
	Pb (29/7)	51%	<0.03	3.10 ± 0.32	3.13 ± 0.32
	Pb (5/9)	49%	0.05 ± 0.02	3.49 ± 0.36	3.54 ± 0.36
	Pb (19/9)	48%	0.06 ± 0.02	2.49 ± 0.31	2.55 ± 0.31
	2006 Pb (13/3)	47%	0.02 ± 0.02	1.33 ± 0.17	1.35 ± 0.17
	Pb (13/3)	47%	0.03 ± 0.02	2.52 ± 0.32	2.55 ± 0.32
	Pb (6/4)	42%	0.06 ± 0.02	1.01 ± 0.13	1.07 ± 0.13
	Pb (4/5)	43%	0.06 ± 0.01	1.30 ± 0.18	1.36 ± 0.18
	Pb (25/7)	53%	0.03 ± 0.01	0.90 ± 0.13	0.93 ± 0.13
	Pb (5/9)	51%	0.02 ± 0.01	0.90 ± 0.12	0.92 ± 0.12
	2007 Pb (7/6)	48%	0.02 ± 0.01	0.87 ± 0.12	0.78 ± 0.12
Sandy	2005 Sa (16/5)	10%	0.07 ± 0.02	2.49 ± 0.26	2.56 ± 0.26
	Sa (16/5)	7%	0.06 ± 0.01	1.96 ± 0.16	2.02 ± 0.16
	Sa (16/5)	9%	0.08 ± 0.02	2.22 ± 0.23	2.30 ± 0.23
	Sa (16/5)	10%	0.10 ± 0.02	2.18 ± 0.16	2.28 ± 0.16
	Sa (20/5)	14%	0.17 ± 0.03	2.87 ± 0.30	3.04 ± 0.30
	Sa (20/5)	9%	0.47 ± 0.06	2.25 ± 0.24	2.72 ± 0.25
	Sa (27/5)	9%	0.06 ± 0.02	3.00 ± 0.31	3.06 ± 0.31
	Sa (27/5)	16%	0.11 ± 0.02	2.11 ± 0.22	2.22 ± 0.22
	Sa (3/6)	4%	0.08 ± 0.02	2.13 ± 0.22	2.21 ± 0.22
	Sa (3/6)	4%	0.27 ± 0.04	2.20 ± 0.23	2.47 ± 0.23
	Sa (17/6)	12%	0.08 ± 0.02	2.94 ± 0.31	3.02 ± 0.31
	Sa (17/6)	10%	0.10 ± 0.02	3.17 ± 0.33	3.27 ± 0.33
	Sa (7/7)	13%	0.04 ± 0.01	1.65 ± 0.21	1.69 ± 0.21
	Sa (29/7)	19%	<0.02	1.91 ± 0.20	1.93 ± 0.20
	Sa (29/7)	15%	<0.02	2.70 ± 0.28	2.72 ± 0.28
	Sa (8/9)	12%	<0.02	1.32 ± 0.14	1.34 ± 0.14
	2006 Sa (13/3)	13%	<0.02	0.40 ± 0.06	0.43 ± 0.06
	Sa (13/3)	14%	<0.02	0.90 ± 0.12	0.92 ± 0.12
	Sa (6/4)	8%	<0.02	0.64 ± 0.08	0.66 ± 0.08
	Sa (4/5)	9%	0.01 ± 0.01	0.63 ± 0.09	0.63 ± 0.09
Sa (25/7)	18%	<0.01	0.45 ± 0.06	0.46 ± 0.06	
Sa (16/8)	13%	<0.01	0.49 ± 0.07	0.50 ± 0.07	
Sa (4/9)	13%	<0.01	0.64 ± 0.09	0.66 ± 0.09	
2005 Sb (16/5)	5%	0.03 ± 0.01	2.48 ± 0.20	2.51 ± 0.20	
Sb (16/5)	6%	0.04 ± 0.01	1.28 ± 0.10	1.32 ± 0.10	
Sb (16/5)	4%	0.03 ± 0.01	0.98 ± 0.11	1.01 ± 0.11	
Sb (16/5)	7%	0.08 ± 0.02	1.21 ± 0.13	1.29 ± 0.13	
Sb (20/5)	13%	0.11 ± 0.18	1.85 ± 0.19	1.96 ± 0.26	
Sb (20/5)	10%	0.27 ± 0.04	2.19 ± 0.23	2.46 ± 0.23	
Sb (27/5)	11%	0.10 ± 0.02	1.47 ± 0.16	1.57 ± 0.16	
Sb (27/5)	9%	0.09 ± 0.02	1.88 ± 0.20	1.97 ± 0.20	
Sb (3/6)	5%	0.11 ± 0.02	2.63 ± 0.28	2.74 ± 0.28	
Sb (3/6)	8%	0.21 ± 0.03	1.30 ± 0.14	1.51 ± 0.14	
Sb (17/6)	10%	0.12 ± 0.02	1.99 ± 0.21	2.11 ± 0.21	
Sb (17/6)	10%	0.02 ± 0.02	1.61 ± 0.20	1.63 ± 0.20	
Sb (17/6)	9%	0.04 ± 0.01	1.76 ± 0.19	1.80 ± 0.19	

Table A1 Measured tritium concentration in lysimeter soils

Soil	Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)		
			Aqueous	OBT	Total
2006	Sb (7/7)	10%	0.03 ± 0.01	1.47 ± 0.19	1.50 ± 0.19
	Sb (29/7)	14%	<0.03	0.90 ± 0.10	0.93 ± 0.10
	Sb (29/7)	12%	0.04 ± 0.02	5.63 ± 0.58	5.67 ± 0.58
	Sb (5/9)	20%	0.02 ± 0.01	0.91 ± 0.10	0.93 ± 0.10
	Sb (19/9)	11%	<0.02	0.69 ± 0.09	0.71 ± 0.09
	Sb (13/3)	16%	0.02 ± 0.02	1.75 ± 0.22	1.77 ± 0.22
	Sb (13/3)	10%	<0.02	1.12 ± 0.14	1.14 ± 0.14
	Sb (6/4)	8%	0.02 ± 0.01	0.36 ± 0.05	0.38 ± 0.05
	Sb (4/5)	8%	<0.01	0.58 ± 0.08	0.59 ± 0.08
	Sb (25/7)	9%	<0.01	0.51 ± 0.07	0.52 ± 0.07
	Sb (5/9)	19%	0.03 ± 0.01	0.75 ± 0.10	0.78 ± 0.10
	2007	Sb (7/6)	11%	0.01 ± 0.01	0.95 ± 0.12

Table A2 Measured tritium concentration in container soils

Soil	Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)			
			Aqueous	OBT	Total	
Treated soil						
Clay	2005	C1 (25/5)	25%	0.51 ± 0.06	4.24 ± 0.44	4.75 ± 0.44
		C1 (4/8)	22%	<0.02	0.86 ± 0.09	0.88 ± 0.09
		C1 (18/8)	15%	0.02 ± 0.01	2.82 ± 0.27	2.84 ± 0.27
	2006	C1 (4/5)	26%	0.04 ± 0.01	0.80 ± 0.11	0.84 ± 0.11
		C1 (8/8)	27%	0.02 ± 0.01	0.62 ± 0.09	0.64 ± 0.09
	2005	C2 (25/5)	26%	0.43 ± 0.05	2.95 ± 0.31	3.38 ± 0.31
		C2 (4/8)	20%	<0.02	0.87 ± 0.09	0.89 ± 0.09
		C2 (18/8)	15%	0.02 ± 0.01	2.63 ± 0.25	2.65 ± 0.25
	2006	C2 (4/5)	26%	0.03 ± 0.01	0.93 ± 0.13	0.96 ± 0.13
		C2 (8/8)	27%	0.03 ± 0.01	0.53 ± 0.08	0.56 ± 0.08
	2005	C4 (25/5)	28%	0.43 ± 0.05	4.39 ± 0.46	4.82 ± 0.46
		C4 (31/8)	16%	<0.02	1.24 ± 0.13	1.26 ± 0.13
	2006	C4 (4/5)	31%	0.04 ± 0.01	0.83 ± 0.12	0.87 ± 0.12
		C4 (16/8)	26%	0.02 ± 0.01	0.71 ± 0.10	0.73 ± 0.10
	2005	C5 (25/5)	28%	0.57 ± 0.07	5.09 ± 0.53	5.66 ± 0.53
		C5 (31/8)	11%	<0.02	0.66 ± 0.07	0.69 ± 0.07
	2006	C5 (4/5)	29%	0.02 ± 0.01	0.90 ± 0.13	0.92 ± 0.13
		C5 (16/8)	25%	0.02 ± 0.01	0.67 ± 0.09	0.68 ± 0.09
	2005	C7 (25/5)	27%	0.41 ± 0.05	2.93 ± 0.31	3.34 ± 0.31
		C7 (5/9)	29%	0.03 ± 0.02	2.42 ± 0.25	2.45 ± 0.25
	2006	C7 (4/5)	29%	0.04 ± 0.01	0.98 ± 0.14	1.02 ± 0.14
		C7 (26/7)	28%	<0.01	0.74 ± 0.10	0.75 ± 0.10
	2005	C8 (25/5)	26%	0.33 ± 0.04	2.67 ± 0.28	3.00 ± 0.28
		C8 (5/9)	23%	0.02 ± 0.01	1.01 ± 0.11	1.03 ± 0.11
2006	C8 (4/5)	28%	0.03 ± 0.01	1.01 ± 0.14	1.04 ± 0.14	

Table A2 Measured tritium concentration in container soils

Soil	Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)		
			Aqueous	OBT	Total
2005	C8 (26/7)	28%	0.01 ± 0.01	0.82 ± 0.11	0.83 ± 0.11
	C10 (25/5)	25%	0.34 ± 0.04	3.58 ± 0.37	3.92 ± 0.37
2006	C10 (19/9)	34%	0.01 ± 0.01	0.58 ± 0.07	0.59 ± 0.07
	C10 (5/9)	34%	0.02 ± 0.01	0.38 ± 0.05	0.40 ± 0.05
2005	C11 (25/5)	27%	0.39 ± 0.05	6.44 ± 0.66	6.83 ± 0.66
	C11 (19/9)	23%	<0.03	0.44 ± 0.06	0.47 ± 0.06
2006	C11 (5/9)	35%	0.01 ± 0.01	0.09 ± 0.02	0.10 ± 0.02
2005	C13 (20/5)	32%	0.40 ± 0.05	7.16 ± 0.74	7.56 ± 0.74
2006	C13 (7/7)	24%	0.02 ± 0.01	0.54 ± 0.08	0.56 ± 0.08
2005	C14 (20/5)	32%	0.14 ± 0.02	1.74 ± 0.18	1.88 ± 0.18
2006	C14 (7/7)	27%	0.02 ± 0.01	0.84 ± 0.12	0.86 ± 0.12
2005	C16 (25/5)	26%	0.22 ± 0.03	3.26 ± 0.34	3.48 ± 0.34
	C16 (8/9)	10%	<0.02	1.81 ± 0.19	1.83 ± 0.19
2006	C16 (13/3)	23%	0.01 ± 0.02	0.97 ± 0.12	0.98 ± 0.12
	C16 (4/5)	27%	0.05 ± 0.01	1.18 ± 0.16	1.23 ± 0.16
2005	C16 (4/9)	35%	0.01 ± 0.01	0.67 ± 0.09	0.68 ± 0.09
	C17 (25/5)	26%	0.16 ± 0.03	1.87 ± 0.20	2.03 ± 0.20
2006	C17 (8/9)	13%	0.02 ± 0.01	1.27 ± 0.14	1.29 ± 0.14
	C17 (4/5)	27%	0.03 ± 0.01	0.73 ± 0.10	0.76 ± 0.10
2005	C17 (4/9)	29%	0.01 ± 0.01	0.61 ± 0.09	0.62 ± 0.09
	L1 (25/5)	21%	0.45 ± 0.06	6.07 ± 0.63	6.52 ± 0.63
2006	L1 (18/8)	11%	<0.02	1.41 ± 0.15	1.43 ± 0.15
	L1 (4/5)	12%	0.02 ± 0.00	0.87 ± 0.12	0.89 ± 0.12
2005	L1 (8/8)	16%	0.02 ± 0.01	0.59 ± 0.08	0.61 ± 0.08
	L2 (25/5)	20%	0.24 ± 0.03	3.78 ± 0.04	4.02 ± 0.05
2006	L2 (4/8)	11%	0.01 ± 0.01	1.87 ± 0.20	1.88 ± 0.20
	L2 (18/8)	16%	0.02 ± 0.02	1.31 ± 0.14	1.33 ± 0.14
2005	L2 (4/5)	11%	0.02 ± 0.00	0.99 ± 0.14	1.01 ± 0.14
	L2 (8/8)	16%	0.02 ± 0.01	0.61 ± 0.09	0.63 ± 0.09
2006	L4 (25/5)	19%	0.16 ± 0.03	2.26 ± 0.02	2.42 ± 0.04
	L4 (31/8)	7%	<0.02	0.85 ± 0.09	0.87 ± 0.09
2005	L4 (4/5)	10%	0.01 ± 0.00	0.83 ± 0.11	0.84 ± 0.11
	L4 (16/8)	18%	0.02 ± 0.01	0.52 ± 0.07	0.53 ± 0.07
2006	L5 (25/5)	20%	0.28 ± 0.02	3.04 ± 0.06	3.32 ± 0.06
	L5 (31/8)	5%	0.03 ± 0.02	2.31 ± 0.24	2.34 ± 0.24
2005	L5 (4/5)	13%	0.02 ± 0.01	0.60 ± 0.08	0.62 ± 0.08
	L5 (16/8)	18%	0.01 ± 0.01	0.53 ± 0.07	0.54 ± 0.07
2006	L7 (25/5)	21%	0.32 ± 0.02	3.61 ± 0.06	3.93 ± 0.07
	L7 (5/9)	20%	<0.02	1.15 ± 0.12	1.17 ± 0.12
2005	L7 (4/5)	11%	0.01 ± 0.00	0.80 ± 0.11	0.81 ± 0.11
	L7 (26/7)	18%	<0.01	0.68 ± 0.10	0.69 ± 0.10
2006	L8 (25/5)	21%	0.26 ± 0.02	3.42 ± 0.06	3.68 ± 0.07
	L8 (5/9)	20%	<0.02	1.37 ± 0.15	1.39 ± 0.15
2005	L8 (4/5)	13%	0.01 ± 0.00	0.76 ± 0.11	0.77 ± 0.11
	L8 (26/7)	17%	<0.01	0.66 ± 0.09	0.67 ± 0.09

Table A2 Measured tritium concentration in container soils

Soil	Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)			
			Aqueous	OBT	Total	
2005	L10 (25/5)	17%	0.23 ± 0.02	2.01 ± 0.05	2.25 ± 0.05	
	L10 (19/9)	28%	<0.02	0.32 ± 0.04	0.34 ± 0.04	
2006	L10 (5/9)	18%	0.01 ± 0.01	0.56 ± 0.08	0.57 ± 0.08	
2005	L11 (25/5)	21%	0.33 ± 0.02	4.56 ± 0.07	4.89 ± 0.08	
	L11 (19/9)	24%	0.03 ± 0.02	0.52 ± 0.07	0.54 ± 0.07	
2006	L11 (5/9)	17%	0.01 ± 0.01	0.54 ± 0.08	0.55 ± 0.08	
2005	L13 (20/5)	21%	0.09 ± 0.02	1.71 ± 0.02	1.80 ± 0.03	
2006	L13 (7/7)	18%	0.01 ± 0.01	0.74 ± 0.10	0.75 ± 0.10	
2005	L14 (20/5)	23%	0.19 ± 0.03	2.48 ± 0.26	2.67 ± 0.26	
2006	L14 (7/7)	19%	0.03 ± 0.01	0.89 ± 0.12	0.92 ± 0.12	
2005	L16 (25/5)	18%	0.21 ± 0.02	1.70 ± 0.04	1.91 ± 0.05	
	L16 (8/9)	14%	0.01 ± 0.01	0.81 ± 0.09	0.82 ± 0.09	
2006	L16 (13/3)	18%	<0.02	0.85 ± 0.11	0.87 ± 0.11	
	L16 (4/5)	12%	0.01 ± 0.01	0.77 ± 0.11	0.78 ± 0.11	
	L16 (4/9)	19%	0.01 ± 0.01	0.42 ± 0.06	0.43 ± 0.06	
2005	L17 (25/5)	22%	0.48 ± 0.07	4.78 ± 0.60	5.26 ± 0.60	
	L17 (8/9)	16%	0.02 ± 0.01	1.37 ± 0.15	1.39 ± 0.15	
2006	L17 (4/5)	13%	<0.01	0.86 ± 0.12	0.87 ± 0.12	
	L17 (4/9)	19%	0.02 ± 0.01	0.59 ± 0.08	0.60 ± 0.08	
Peat	2005	P1 (25/5)	42%	0.61 ± 0.08	5.53 ± 0.69	6.14 ± 0.69
		P1 (18/8)	44%	0.04 ± 0.02	2.34 ± 0.23	2.38 ± 0.23
2006	P1 (4/5)	40%	0.10 ± 0.02	2.47 ± 0.34	2.57 ± 0.34	
	P1 (8/8)	47%	0.06 ± 0.01	1.91 ± 0.26	1.97 ± 0.26	
2005	P2 (25/5)	40%	0.62 ± 0.08	4.35 ± 0.54	4.97 ± 0.55	
	P2 (18/8)	43%	0.02 ± 0.02	3.25 ± 0.31	3.27 ± 0.31	
2006	P2 (4/5)	38%	0.05 ± 0.01	1.95 ± 0.27	2.00 ± 0.27	
	P2 (8/8)	46%	0.02 ± 0.01	1.08 ± 0.15	1.10 ± 0.15	
2005	P4 (25/5)	41%	<0.03	1.23 ± 0.13	1.26 ± 0.13	
	P4 (31/8)	26%	0.08 ± 0.02	4.82 ± 0.50	4.90 ± 0.50	
2006	P4 (4/5)	40%	0.07 ± 0.01	2.21 ± 0.30	2.28 ± 0.30	
	P4 (16/8)	41%	0.02 ± 0.01	0.84 ± 0.12	0.86 ± 0.12	
2005	P5 (25/5)	40%	0.49 ± 0.07	4.30 ± 0.54	4.79 ± 0.54	
	P5 (31/8)	23%	0.05 ± 0.02	4.79 ± 0.50	4.84 ± 0.50	
2006	P5 (4/5)	38%	0.04 ± 0.01	1.38 ± 0.19	1.42 ± 0.19	
	P5 (16/8)	41%	0.02 ± 0.01	0.85 ± 0.12	0.87 ± 0.12	
2005	P7 (25/5)	39%	0.37 ± 0.05	3.54 ± 0.44	3.91 ± 0.44	
	P7 (5/9)	44%	<0.03	2.32 ± 0.24	2.35 ± 0.24	
2006	P7 (4/5)	41%	0.08 ± 0.02	1.92 ± 0.27	2.00 ± 0.27	
	P7 (26/7)	51%	0.05 ± 0.01	1.76 ± 0.24	1.81 ± 0.24	
2005	P8 (25/5)	39%	<0.02	3.15 ± 0.33	3.17 ± 0.33	
	P8 (5/9)	33%	0.02 ± 0.02	3.82 ± 0.40	3.84 ± 0.40	
2006	P8 (4/5)	38%	0.06 ± 0.01	2.33 ± 0.32	2.39 ± 0.32	
	P8 (26/7)	47%	0.02 ± 0.01	1.44 ± 0.20	1.46 ± 0.20	
2005	P10 (25/5)	46%	<0.02	1.30 ± 0.14	1.32 ± 0.14	
	P10 (19/9)	47%	0.05 ± 0.02	1.85 ± 0.19	1.90 ± 0.19	

Table A2 Measured tritium concentration in container soils

Soil	Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)		
			Aqueous	OBT	Total
	2006 P10 (5/9)	58%	0.02 ± 0.01	1.60 ± 0.22	1.62 ± 0.22
	2005 P11 (25/5)	38%	0.67 ± 0.09	8.90 ± 1.10	9.57 ± 1.10
	P11 (19/9)	46%	<0.03	1.18 ± 0.13	1.21 ± 0.13
	2006 P11 (5/9)	44%	0.02 ± 0.01	1.88 ± 0.26	1.90 ± 0.26
	2005 P13 (20/5)	36%	0.32 ± 0.04	5.90 ± 0.61	6.22 ± 0.61
	2006 P13 (7/7)	42%	0.04 ± 0.01	1.55 ± 0.21	1.59 ± 0.21
	2005 P14 (20/5)	38%	0.26 ± 0.04	3.20 ± 0.33	3.46 ± 0.33
	2006 P14 (7/7)	43%	0.04 ± 0.01	1.26 ± 0.18	1.30 ± 0.18
	2005 P16 (25/5)	36%	0.61 ± 0.08	6.58 ± 0.82	7.19 ± 0.82
	P16 (8/9)	35%	0.10 ± 0.02	4.14 ± 0.43	4.24 ± 0.43
	2006 P16 (13/3)	44%	0.03 ± 0.02	1.29 ± 0.16	1.32 ± 0.16
	P16 (4/5)	36%	0.05 ± 0.01	1.50 ± 0.21	1.55 ± 0.21
	P16 (4/9)	48%	<0.01	0.89 ± 0.12	0.90 ± 0.12
	2005 P17 (25/5)	41%	<0.02	0.52 ± 0.06	0.55 ± 0.06
	P17 (8/9)	39%	0.07 ± 0.02	3.17 ± 0.33	3.24 ± 0.33
	2006 P17 (4/5)	41%	0.05 ± 0.01	1.26 ± 0.17	1.31 ± 0.17
	P17 (4/9)	52%	0.04 ± 0.01	1.04 ± 0.14	1.08 ± 0.14
Sandy	2005 S1 (25/5)	16%	0.04 ± 0.02	3.84 ± 0.40	3.88 ± 0.40
	S1 (18/8)	18%	0.02 ± 0.01	1.31 ± 0.13	1.33 ± 0.13
	2006 S1 (4/5)	12%	0.02 ± 0.00	0.81 ± 0.11	0.83 ± 0.11
	S1 (8/8)	11%	0.01 ± 0.01	0.60 ± 0.09	0.62 ± 0.09
	2005 S2 (25/5)	16%	0.23 ± 0.04	3.12 ± 0.39	3.35 ± 0.39
	2006 S2 (4/5)	12%	0.01 ± 0.00	0.61 ± 0.09	0.62 ± 0.09
	S2 (8/8)	11%	0.01 ± 0.01	0.42 ± 0.06	0.43 ± 0.06
	2005 S4 (25/5)	19%	0.22 ± 0.04	3.49 ± 0.44	3.71 ± 0.44
	S4 (31/8)	6%	0.01 ± 0.01	1.51 ± 0.16	1.52 ± 0.16
	2006 S4 (4/5)	13%	0.02 ± 0.00	0.52 ± 0.07	0.54 ± 0.07
	S4 (16/8)	11%	0.01 ± 0.01	0.47 ± 0.07	0.48 ± 0.07
	2005 S5 (25/5)	17%	<0.02	2.35 ± 0.25	2.37 ± 0.25
	S5 (31/8)	6%	<0.02	1.15 ± 0.12	1.17 ± 0.12
	2006 S5 (4/5)	8%	0.01 ± 0.00	0.80 ± 0.11	0.81 ± 0.11
	S5 (16/8)	13%	<0.01	0.64 ± 0.09	0.65 ± 0.09
	2005 S7 (25/5)	16%	0.08 ± 0.02	2.79 ± 0.35	2.87 ± 0.35
	S7 (5/9)	13%	<0.02	0.70 ± 0.08	0.73 ± 0.08
	2006 S7 (4/5)	12%	0.02 ± 0.00	0.54 ± 0.08	0.56 ± 0.08
	S7 (26/7)	13%	<0.01	0.33 ± 0.05	0.34 ± 0.05
	2005 S8 (25/5)	17%	0.14 ± 0.03	2.94 ± 0.37	3.08 ± 0.37
	S8 (5/9)	9%	<0.02	1.12 ± 0.12	1.14 ± 0.12
	2006 S8 (4/5)	10%	0.02 ± 0.00	0.74 ± 0.10	0.76 ± 0.10
	S8 (26/7)	16%	0.01 ± 0.01	0.50 ± 0.07	0.51 ± 0.07
	2005 S10 (25/5)	18%	0.23 ± 0.04	3.37 ± 0.42	3.60 ± 0.42
	S10 (19/9)	32%	0.01 ± 0.01	0.97 ± 0.11	0.98 ± 0.11
	2006 S10 (5/9)	11%	0.01 ± 0.01	1.05 ± 0.15	1.06 ± 0.15
	2005 S11 (25/5)	18%	0.25 ± 0.04	2.58 ± 0.32	2.83 ± 0.32
	S11 (19/9)	23%	<0.02	0.77 ± 0.09	0.80 ± 0.09

Table A2 Measured tritium concentration in container soils

Soil	Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)			
			Aqueous	OBT	Total	
	2006	S11 (5/9)	14%	0.01 ± 0.01	0.70 ± 0.10	0.71 ± 0.10
	2005	S13 (20/5)	19%	0.12 ± 0.02	3.17 ± 0.33	3.29 ± 0.33
	2006	S13 (7/7)	15%	0.01 ± 0.01	0.71 ± 0.10	0.72 ± 0.10
	2005	S14 (20/5)	19%	0.22 ± 0.03	5.00 ± 0.52	5.22 ± 0.52
	2006	S14 (7/7)	14%	0.00 ± 0.01	0.88 ± 0.12	0.88 ± 0.12
	2005	S16 (25/5)	16%	0.25 ± 0.04	4.35 ± 0.54	4.60 ± 0.54
		S16 (8/9)	7%	<0.02	1.26 ± 0.13	1.28 ± 0.13
	2006	S16 (13/3)	16%	<0.02	0.73 ± 0.10	0.75 ± 0.10
		S16 (4/5)	10%	0.02 ± 0.01	0.70 ± 0.10	0.72 ± 0.10
		S16 (4/9)	14%	0.02 ± 0.01	0.45 ± 0.06	0.46 ± 0.06
	2005	S17 (25/5)	17%	0.13 ± 0.02	1.80 ± 0.23	1.93 ± 0.23
		S17 (8/9)	8%	0.01 ± 0.01	1.16 ± 0.12	1.17 ± 0.12
	2006	S17 (4/5)	9%	0.00 ± 0.01	0.46 ± 0.07	0.46 ± 0.07
		S17 (4/9)	17%	<0.01	0.30 ± 0.04	0.31 ± 0.04
Untreated (Control) soil						
Clay	2005	C3 (4/8)	14%	<0.02	0.06 ± 0.01	0.08 ± 0.01
		C3 (18/8)	12%	<0.02	0.03 ± 0.01	0.05 ± 0.01
	2006	C3 (4/5)	21%	<0.01	<0.009	<0.019
		C3 (8/8)	22%	<0.01	<0.01	<0.02
	2005	C6 (31/8)	12%	<0.02	<0.02	<0.04
	2006	C6 (4/5)	19%	<0.01	<0.009	<0.019
	2005	C9 (5/9)	24%	<0.02	<0.02	<0.04
	2006	C9 (4/5)	16%	<0.01	<0.009	<0.019
		C9 (26/7)	27%	<0.01	0.006 ± 0.006	0.016 ± 0.006
	2005	C12 (19/9)	22%	0.05 ± 0.02	<0.02	0.07 ± 0.02
	2006	C12 (5/9)	33%	<0.01	<0.009	<0.02
	2005	C15 (20/5)	31%	<0.02	<0.02	<0.04
	2006	C15 (7/7)	26%	<0.009	0.011 ± 0.005	0.02 ± 0.005
	2005	C18 (8/9)	13%	<0.02	0.01 ± 0.01	0.03 ± 0.01
	2006	C18 (4/5)	18%	<0.01	<0.009	<0.019
		C18 (4/9)	35%	<0.009	0.008 ± 0.005	0.017 ± 0.005
	Loam	2005	L3 (4/8)	17%	<0.02	<0.02
		L3 (18/8)	18%	<0.02	<0.02	<0.04
2006		L3 (4/5)	10%	<0.01	<0.01	<0.02
		L3 (8/8)	15%	<0.01	<0.01	<0.02
2005		L6 (31/8)	6%	<0.02	<0.02	<0.04
2006		L6 (4/5)	10%	<0.01	<0.01	<0.02
		L6 (16/8)	16%	<0.008	0.005 ± 0.005	0.013 ± 0.005
2005		L9 (5/9)	19%	<0.02	<0.02	<0.04
2006		L9 (4/5)	12%	<0.01	<0.01	<0.02
		L9 (26/7)	18%	<0.009	0.005 ± 0.005	0.014 ± 0.005
2005		L12 (19/9)	18%	0.02 ± 0.02	<0.02	0.04 ± 0.02
2006		L12 (5/9)	19%	<0.01	<0.009	<0.019
2005		L15 (20/5)	20%	<0.02	<0.02	<0.04
2005		L18 (8/9)	15%	<0.02	<0.02	<0.04

Table A2 Measured tritium concentration in container soils

Soil		Container (date of sample)	Percent water	Tritium concentration (Bq g ⁻¹)		
				Aqueous	OBT	Total
	2006	L18 (4/5)	10%	<0.01	<0.01	<0.02
		L18 (4/9)	19%	<0.009	<0.007	<0.016
Peat	2005	P3 (4/8)	39%	<0.02	0.01 ± 0.01	0.03 ± 0.01
		P3 (18/8)	40%	<0.02	<0.02	<0.04
	2006	P3 (4/5)	32%	<0.009	0.007 ± 0.005	0.016 ± 0.005
		P3 (8/8)	43%	<0.01	0.005 ± 0.006	0.015 ± 0.006
	2005	P6 (31/8)	22%	0.01 ± 0.01	0.02 ± 0.01	0.03 ± 0.02
	2006	P6 (4/5)	33%	0.005 ± 0.006	0.007 ± 0.005	0.012 ± 0.011
		P6 (16/8)	43%	<0.01	<0.01	<0.02
	2005	P9 (5/9)	40%	<0.03	<0.02	<0.05
	2006	P9 (4/5)	33%	<0.009	0.007 ± 0.005	0.016 ± 0.005
		P9 (26/7)	45%	0.006 ± 0.007	<0.009	0.015 ± 0.007
	2005	P12 (19/9)	43%	<0.03	<0.02	<0.05
	2006	P12 (5/9)	43%	<0.01	<0.009	<0.019
	2005	P15 (20/5)	37%	<0.02	<0.02	<0.04
	2006	P15 (7/7)	38%	0.01 ± 0.006	0.005 ± 0.005	0.015 ± 0.011
	2005	P18 (8/9)	32%	<0.03	<0.02	<0.05
	2006	P18 (4/5)	31%	<0.008	0.005 ± 0.005	0.013 ± 0.005
	P18 (4/9)	44%	<0.01	0.005 ± 0.005	0.015 ± 0.005	
Sandy	2005	S3 (4/8)	8%	<0.02	<0.02	<0.04
		S3 (18/8)	14%	<0.02	<0.02	<0.04
	2006	S3 (4/5)	7%	<0.01	<0.01	<0.02
		S3 (8/8)	8%	<0.01	<0.01	<0.02
	2005	S6 (31/8)	2%	<0.02	<0.02	<0.04
	2006	S6 (4/5)	8%	<0.01	<0.01	<0.02
		S6 (16/8)	9%	<0.009	0.005 ± 0.005	0.014 ± 0.005
	2005	S9 (5/9)	11%	<0.02	<0.02	<0.04
	2006	S9 (4/5)	8%	<0.008	<0.007	<0.015
		S9 (26/7)	11%	<0.009	<0.009	<0.018
	2005	S12 (19/9)	10%	<0.02	<0.02	<0.04
	2006	S12 (5/9)	15%	<0.01	<0.009	<0.019
	2005	S15 (20/5)	83%	<0.02	<0.01	<0.03
	2006	S15 (7/7)	19%	<0.009	0.006 ± 0.005	0.015 ± 0.005
	2005	S18 (8/9)	9%	<0.02	<0.02	<0.04
	2006	S18 (4/5)	7%	<0.007	<0.007	<0.014
	S18 (4/9)	25%	<0.009	0.007 ± 0.005	0.016 ± 0.005	

A2 TRITIUM IN LEACHATE AND RAINWATER

Table A3 Concentrations and fluxes of tritium in leachate

Dates	Volume ml	Tritium content		Dates	Volume ml	Tritium content	
		Bq.ml ⁻¹	Bq total			Bq.ml ⁻¹	Bq total
Clay Soil				Loam Soil			
2005				2005			
18/05 to 27/05	969	0.038 ± 0.006	37 ± 6	18/05 to 27/05	412	0.006 ± 0.002	2.6 ± 1.0
25/05 to 02/06	811	0.108 ± 0.007	88 ± 6	25/05 to 02/06	429	0.166 ± 0.009	71 ± 4
02/06 to 09/06	1,364	0.169 ± 0.007	231 ± 10	02/06 to 09/06	1,233	0.178 ± 0.008	219 ± 10
09/06 to 17/06	2,410	0.22 ± 0.01	530 ± 30	09/06 to 17/06	1,566	0.18 ± 0.01	290 ± 20
17/06 to 27/06	10,200	0.20 ± 0.01	2000 ± 100	17/06 to 27/06	7,066	0.140 ± 0.009	990 ± 60
27/06 to 29/06 ¹	6,500	0.17 ± 0.01	1120 ± 70	27/06 to 29/06 ¹	5,100	0.17 ± 0.01	880 ± 50
29/06 to 12/07	3,600	0.084 ± 0.006	300 ± 20	29/06 to 19/07	2,600	0.066 ± 0.005	170 ± 10
12/07 to 19/07	4,400	0.044 ± 0.004	190 ± 20	19/07 to 26/07	9,900	0.005 ± 0.002	50 ± 20
19/07 to 26/07	9,700	0.002 ± 0.002	20 ± 20	26/07 to 03/08	10,800	0.009 ± 0.003	100 ± 30
26/07 to 03/08	10,200	0.011 ± 0.003	110 ± 30	03/08 to 11/08	10,500	0.013 ± 0.003	140 ± 30
03/08 to 11/08	10,300	0.014 ± 0.003	140 ± 30	11/08 to 23/08	3,200	0.015 ± 0.003	48 ± 9
11/08 to 23/08	3,500	0.017 ± 0.003	60 ± 10	23/08 to 08/09	3,400	0.008 ± 0.002	26 ± 8
23/08 to 08/09	6,300	0.014 ± 0.003	90 ± 20	08/09 to 21/09	9,500	0.016 ± 0.003	160 ± 30
08/09 to 21/09	8,000	0.015 ± 0.003	120 ± 20	21/09 to 01/11	5,000	0.034 ± 0.004	170 ± 20
21/09 to 01/11	2,300	0.022 ± 0.004	51 ± 8				
Total in season 5100 ± 400 Bq ≅ 23% of added tritium				Total in season 3300 ± 300 Bq ≅ 25% of added tritium			
2006				2006			
04/05 to 02/06	2,900	0.033 ± 0.005	90 ± 10	04/05 to 02/06	2,800	0.047 ± 0.006	130 ± 20
02/06 to 24/07	700	0.009 ± 0.003	6 ± 2	No further samples available			
24/07 to 06/09	3,500	0.035 ± 0.005	120 ± 2				
Total in season 224 ± 22 Bq ≅ 1% of added tritium				Total in season 130 ± 20 Bq ≅ 1% of added tritium			
Peat Soil				Sandy Soil			
2005				2005			
18/05 to 27/05	226	0.004 ± 0.003	0.9 ± 0.7	18/05 to 18/05 ²	1,400	0.16 ± 0.02	230 ± 20
25/05 to 02/06	336	0.116 ± 0.006	39 ± 2	18/05 to 27/05	1,437	0.14 ± 0.02	200 ± 20
02/06 to 09/06	449	0.069 ± 0.007	31 ± 3	25/05 to 02/06	1,134	0.28 ± 0.02	310 ± 20
09/06 to 17/06	1,229	0.069 ± 0.007	85 ± 9	02/06 to 09/06	1,333	0.36 ± 0.02	480 ± 20
17/06 to 27/06	6,813	0.076 ± 0.006	520 ± 40	09/06 to 17/06	1,563	0.36 ± 0.02	560 ± 30
27/06 to 29/06 ¹	5,500	0.122 ± 0.007	670 ± 40	17/06 to 27/06	6,777	0.16 ± 0.01	1070 ± 70
29/06 to 19/07	1,800	0.069 ± 0.005	125 ± 9	27/06 to 29/06 ¹	5,800	0.18 ± 0.01	1040 ± 60
19/07 to 26/07	7,800	0.003 ± 0.002	20 ± 20	29/06 to 19/07	1,200	0.069 ± 0.005	83 ± 6
26/07 to 03/08	10,900	0.008 ± 0.003	90 ± 30	19/07 to 26/07	6,900	0.019 ± 0.003	130 ± 20
03/08 to 11/08	10,000	0.011 ± 0.003	110 ± 30	26/07 to 03/08	7,500	0.010 ± 0.003	80 ± 20
11/08 to 23/08	3,200	0.012 ± 0.002	38 ± 6	03/08 to 11/08	9,500	0.014 ± 0.003	130 ± 30
23/08 to 08/09	3,400	0.009 ± 0.003	32 ± 9	11/08 to 23/08	4,500	0.016 ± 0.003	70 ± 10
08/09 to 21/09	10,500	0.010 ± 0.003	100 ± 30	23/08 to 08/09	4,200	0.014 ± 0.003	60 ± 10
21/09 to 01/11	2,800	0.014 ± 0.003	40 ± 8	08/09 to 21/09	4,500	0.015 ± 0.003	70 ± 10
Total in season 1900 ± 200 Bq ≅ 9% of added tritium				Total in season 4600 ± 400 Bq ≅ 21% of added tritium			

Table A3 Concentrations and fluxes of tritium in leachate

Dates	Volume ml	Tritium content		Dates	Volume ml	Tritium content	
		Bq.ml ⁻¹	Bq total			Bq.ml ⁻¹	Bq total
Clay Soil				Loam Soil			
2006				2006			
04/05 to 02/06	2,800	0.03 ± 0.004	90 ± 10	04/05 to 02/06	1,900	0.034 ± 0.005	66 ± 9
02/06 to 24/07	2,500	0.021 ± 0.004	54 ± 10	No further samples available			
24/07 to 06/09	6,200	0.017 ± 0.003	100 ± 20				
Total in season 243 ± 25 Bq ≅ 1% of added tritium				Total in season 66 ± 9 Bq ≅ 1% of added tritium			

¹Thunderstorms overnight on 28/6, ²This container inadvertently badly over watered on 18/5/05.

Table A4 Concentration of tritium in rainwater

Dates	Volume, ml	Tritium content, Bq ml ⁻¹
2005		
16/05 – 27/05	1,644	0.010 ± 0.003
25/05 – 17/06	364	0.012 ± 0.003
17/06 – 27/06	1,210	0.033 ± 0.008
27/06 – 07/07	2,730	<0.004
07/07 – 26/07	1,500	0.003 ± 0.003
26/07 – 08/09	2,394	0.031 ± 0.004
08/09 – 20/09	1,138	0.006 ± 0.002
20/09 – 01/11	5,000	0.006 ± 0.002
2006		
04/05 – 02/06	5,000	0.008 ± 0.003
02/06 – 24/07	1,800	0.007 ± 0.003
24/07 – 06/09	4,500	0.014 ± 0.003

Rainwater was collected using a 0.3 m diameter circular funnel.

A3 TRITIUM IN CROPS

Table A5 Measured concentrations of tritium in cabbage

Soil		Container (date of harvest)	Tritium concentration (Bq g ⁻¹)	
			Aqueous	OBT
Clay	2005	C4 (31/8)	0.03 ± 0.01	0.006 ± 0.005
		C5 (31/8)	0.04 ± 0.01	<0.01
		C6 Blank (31/8)	0.018 ± 0.010	<0.01
	2006	C16 (4/9)	<0.007	<0.006
		C17 (4/9)	<0.004	<0.005
		C18 Blank (4/9) ¹	<0.007	<0.005
Loam	2005	L4 (31/8) ¹	0.06 ± 0.01	0.014 ± 0.006
		L5 (31/8)	0.04 ± 0.01	<0.01
		L6 Blank (31/8)	<0.02	<0.01
	2006	L16 (4/9)	0.015 ± 0.005	0.006 ± 0.003
		L17 (4/9)	0.006 ± 0.004	<0.004
		Lysimeter La (4/9)	0.033 ± 0.009	0.003 ± 0.002
	L18 Blank (4/9)	0.019 ± 0.005	<0.004	
Peat	2005	P4 (31/8)	0.02 ± 0.01	<0.01
		P5 (31/8)	0.03 ± 0.01	<0.01
		P6 Blank (31/8)	<0.02	<0.01
	2006	P16 (4/9)	<0.008	<0.005
		P17 (4/9)	<0.008	<0.006
		Lysimeter Pa (4/9)	0.032 ± 0.006	0.002 ± 0.002
	P18 Blank (4/9)	<0.007	<0.006	
Sandy	2005	S4 (31/8)	0.06 ± 0.01	<0.01
		S5 (31/8)	0.04 ± 0.01	<0.01
		S6 Blank (31/8)	<0.02	<0.01
	2006	S16 (4/9) ¹	0.010 ± 0.004	0.002 ± 0.003
		S17 (4/9) ¹	0.010 ± 0.004	<0.004
		Lysimeter Sa (4/9)	0.048 ± 0.008	0.002 ± 0.002
	S18 Blank (4/9) ¹	<0.005	<0.004	

¹Very small cabbage with no hearts.

Table A6 Measured concentrations of tritium in carrot

Soil		Container (date of harvest)	Tritium concentration (Bq g ⁻¹)			
			Aqueous	OBT		
Clay	2005	C7 (5/9)	0.02 ± 0.01	<0.01		
		C8 (5/9)	0.03 ± 0.01	<0.01		
		C9 Blank (5/9)	<0.02	<0.01		
	2006	C4 (16/8)	0.032 ± 0.006	0.007 ± 0.003		
		C5 (16/8)	0.030 ± 0.006	0.004 ± 0.002		
		C6 Blank	Crop failed			
Loam	2005	L7 (5/9)	0.02 ± 0.01	0.011 ± 0.006		
		L8 (5/9)	0.011 ± 0.009	0.013 ± 0.008		
		Lysimeter Lb (5/9)	0.10 ± 0.02	0.024 ± 0.007		
		L9 Blank (5/9)	<0.02	<0.01		
	2006	L4 (16/8)	0.030 ± 0.006	0.007 ± 0.003		
		L5 (16/8)	0.025 ± 0.006	0.007 ± 0.003		
		Lysimeter La (16/8)	0.041 ± 0.008	<0.0005		
		L6 Blank (16/8)	0.016 ± 0.005	0.006 ± 0.003		
		Peat	2005	P7 (5/9)	<0.02	<0.01
				P8 (5/9)	0.04 ± 0.01	0.009 ± 0.006
Lysimeter Pb (5/9)	0.12 ± 0.02			<0.01		
P9 Blank (5/9)	<0.01			<0.01		
2006	P4 (16/8)		0.027 ± 0.006	0.009 ± 0.003		
	P5 (16/8)		0.020 ± 0.005	0.003 ± 0.002		
Sandy	2005	S7 (5/9)	0.07 ± 0.01	<0.01		
		S8 (5/9)	0.03 ± 0.01	<0.01		
		Lysimeter Sb (5/9)	0.11 ± 0.02	0.009 ± 0.005		
		S9 Blank (5/9)	<0.02	<0.01		
	2006	S4 (16/8)	0.032 ± 0.007	0.007 ± 0.007		
		S5 (16/8)	0.024 ± 0.006	0.005 ± 0.004		
		Lysimeter Sa (16/8)	0.020 ± 0.005	0.004 ± 0.003		
		S6 Blank (16/8)	0.016 ± 0.005	<0.006		

Table A7 Measured concentrations of tritium in dwarf French beans

Soil		Container (date of harvest)	Tritium concentration (Bq g ⁻¹)			
			Aqueous	OBT		
Clay	2005	C1 (4/8)	0.02 ± 0.02	<0.02		
		C1 (18/8)	0.04 ± 0.02	<0.02		
		C2 (4/8)	0.03 ± 0.02	<0.02		
		C2 (18/8)	0.07 ± 0.02	<0.02		
		C3 Blank (4/8)	<0.02	<0.02		
		C3 Blank(18/8)	0.07 ± 0.01	0.008 ± 0.006		
	2006	C7 (17/7)	0.025 ± 0.006	0.004 ± 0.003		
		C7 (26/7)	0.020 ± 0.005	<0.007		
		C8 (17/7)	0.024 ± 0.006	0.004 ± 0.002		
		C8 (26/7)	0.023 ± 0.006	<0.004		
		C9 Blank (17/7)	0.014 ± 0.004	0.003 ± 0.002		
		C9 Blank (26/7)	0.019 ± 0.005	<0.005		
		Loam	2005	L1 (18/8)	0.05 ± 0.01	0.11 ± 0.006
				L2 (4/8)	0.03 ± 0.02	0.008 ± 0.010
L2 (18/8)	0.06 ± 0.01			0.006 ± 0.005		
L3 Blank (4/8)	<0.02			<0.02		
L3 Blank(18/8)	0.05 ± 0.01			0.016 ± 0.006		
2006	L7 (17/7)		0.029 ± 0.005	0.008 ± 0.003		
	L7 (26/7)		0.029 ± 0.006	<0.006		
	L8 (17/7)		0.030 ± 0.006	<0.004		
	L8 (26/7)		0.026 ± 0.006	<0.005		
	Lysimeter La (17/7)		0.06 ± 0.01	0.003 ± 0.003		
	Lysimeter La (25/7)		0.039 ± 0.007	0.003 ± 0.003		
	L9 Blank (17/7)		0.014 ± 0.004	0.002 ± 0.002		
	L9 Blank (26/7)		0.019 ± 0.005	<0.006		
	Peat		2005	P1 (18/8)	0.06 ± 0.02	<0.02
P2 (18/8)		0.04 ± 0.02		<0.02		
P3 Blank (4/8)		<0.03		<0.02		
P3 Blank(18/8)		0.06 ± 0.01		<0.01		
2006		P7 (17/7)	0.021 ± 0.005	0.004 ± 0.002		
		P7 (26/7)	0.029 ± 0.006	0.002 ± 0.002		
		P8 (17/7)	0.021 ± 0.005	<0.004		
		P8 (26/7)	0.021 ± 0.005	0.007 ± 0.002		
		Lysimeter Pa (17/7)	0.049 ± 0.009	0.004 ± 0.003		
		Lysimeter Pa (25/7)	0.028 ± 0.006	0.005 ± 0.003		
		P9 Blank (17/7)	0.014 ± 0.005	0.003 ± 0.003		
		P9 Blank (26/7)	0.024 ± 0.005	0.004 ± 0.002		
		Sandy	2005	S1 (18/8)	0.05 ± 0.02	<0.02
				S3 Blank (4/8)	<0.02	<0.02
S3 Blank(12/8)	0.05 ± 0.01			<0.02		
2006	S7 (17/7)		0.036 ± 0.007	0.005 ± 0.003		
	S7 (26/7)		0.030 ± 0.006	0.007 ± 0.003		
	S8 (17/7)		0.036 ± 0.007	0.006 ± 0.003		
	S8 (26/7)		0.029 ± 0.006	0.007 ± 0.003		
	Lysimeter Sa (17/7)		0.045 ± 0.008	0.005 ± 0.003		
	Lysimeter Sa (25/7)		0.029 ± 0.006	0.004 ± 0.003		
	S9 Blank (17/7)		0.012 ± 0.004	0.003 ± 0.003		

Table A7 Measured concentrations of tritium in dwarf French beans

Soil	Container (date of harvest)	Tritium concentration (Bq g ⁻¹)	
		Aqueous	OBT
	S9 Blank (26/7)	0.018 ± 0.004	0.002 ± 0.002

Table A8 Measured tritium concentrations in potatoes

Soil		Container (date of harvest)	Tritium concentration (Bq g ⁻¹)	
			Aqueous	OBT
Clay	2005	C16 (8/9)	0.05 ± 0.01	<0.01
		C17 (8/9)	0.03 ± 0.01	<0.01
		C18 Blank (8/9)	<0.02	<0.01
	2006	C1 (8/8)	0.021 ± 0.005	<0.005
		C2 (8/8)	0.015 ± 0.005	0.004 ± 0.003
		C3 Blank (8/8)	<0.006	0.004 ± 0.003
Loam	2005	L16 (8/9)	0.04 ± 0.01	0.019 ± 0.007
		L17 (8/9)	0.03 ± 0.01	<0.01
		Lysimeter La (8/9)	0.13 ± 0.02	0.017 ± 0.007
		<i>Duplicate analysis</i>	0.12 ± 0.02	0.033 ± 0.008
		<i>Duplicate analysis</i>	0.11 ± 0.02	0.023 ± 0.008
		L18 Blank (8/9)	<0.02	<0.01
	2006	L1 (8/8)	0.021 ± 0.006	0.005 ± 0.004
		L2 (8/8)	0.011 ± 0.004	<0.006
		Lysimeter Lb (25/7)	0.024 ± 0.005	0.007 ± 0.003
		L3 Blank (8/8)	<0.006	<0.006
Peat	2005	P16 (8/9)	0.04 ± 0.01	0.009 ± 0.007
		P17 (8/9)	0.06 ± 0.01	0.018 ± 0.007
		Lysimeter Pa (8/9)	0.07 ± 0.01	0.013 ± 0.006
		<i>Duplicate analysis</i>	0.09 ± 0.02	0.012 ± 0.006
		(Flesh)	0.08 ± 0.02	0.012 ± 0.005
		(Peel)	0.09 ± 0.02	0.015 ± 0.006
	P18 Blank (8/9)	0.01 ± 0.01	0.08 ± 0.01	
	2006	P1 (8/8)	0.027 ± 0.006	0.009 ± 0.003
		P2 (8/8)	0.026 ± 0.005	0.016 ± 0.004
		Lysimeter Pb (25/7)	0.057 ± 0.010	0.006 ± 0.003
P3 Blank (8/8)		0.010 ± 0.004	0.016 ± 0.004	
Sandy	2005	S16 (8/9)	0.06 ± 0.01	0.10 ± 0.005
		S17 (8/9)	0.08 ± 0.01	0.013 ± 0.004
		Lysimeter Sa (8/9)	0.20 ± 0.02	0.029 ± 0.008
		<i>Duplicate analysis</i>	0.20 ± 0.03	0.029 ± 0.007
		S18 Blank (8/9)	0.018 ± 0.007	<0.007
	2006	S1 (8/8)	0.034 ± 0.006	0.014 ± 0.003
		S2 (8/8)	0.031 ± 0.006	0.005 ± 0.002
		Lysimeter Sb (25/7)	0.040 ± 0.007	0.011 ± 0.003
		S3 Blank (8/8)	0.011 ± 0.004	0.007 ± 0.003

Table A9 Measured tritium concentrations in strawberries

Soil		Container (date of harvest)	Tritium concentration (Bq g ⁻¹)	
			Aqueous	OBT
Clay	2005	Crop failure		
	2006	C13 (19/6)	0.012 ± 0.005	<0.004
		C13 (7/7)	0.014 ± 0.005	<0.005
		C14 (19/6)	0.021 ± 0.005	<0.004
		C14 (7/7)	0.010 ± 0.004	<0.005
		C15 Blank (19/6)	0.010 ± 0.004	<0.003
		C15 Blank (7/7)	0.007 ± 0.004	<0.005
Loam	2005	Crop failure		
	2006	L13 (19/6)	0.017 ± 0.005	<0.004
		L13 (7/7)	0.014 ± 0.005	<0.004
		L14 (19/6)	0.018 ± 0.005	<0.004
		L14 (4/7)	0.012 ± 0.004	<0.004
		L15 Blank (19/6)	0.011 ± 0.004	<0.004
		L15 Blank (7/7)	0.012 ± 0.004	0.003 ± 0.002
Peat	2005	Crop failure		
	2006	P13 (19/6)	0.014 ± 0.005	<0.004
		P13 (7/7)	0.017 ± 0.005	0.002 ± 0.002
		P14 (19/6)	0.020 ± 0.006	<0.003
		P14 (7/7)	0.016 ± 0.004	0.004 ± 0.002
		P15 Blank (19/6)	0.017 ± 0.005	<0.004
		P15 Blank (7/7)	0.017 ± 0.004	0.002 ± 0.002
Sandy	2005	Crop failure		
	2006	S13 (20/6)	0.016 ± 0.005	<0.004
		S13 (7/7)	0.010 ± 0.004	<0.005
		S14 (19/6)	0.018 ± 0.005	<0.004
		S14 (7/7)	0.016 ± 0.004	0.003 ± 0.002
		S15 Blank (19/6)	0.012 ± 0.005	<0.004
		S15 Blank (7/7)	0.016 ± 0.004	0.003 ± 0.003

A4 TRITIUM IN GRASS

Table A10 Measured concentrations of tritium in grass

Soil	Container	Date	Tritium concentration (Bq g ⁻¹)		
			Aqueous	OBT	
Clay	C10	2005	27 Jun	0.08 ± 0.02	0.06 ± 0.01
			12 Jul	0.06 ± 0.01	0.04 ± 0.01
			26 Jul	<0.02	0.07 ± 0.01
			23 Aug	0.08 ± 0.02	0.018 ± 0.008
			14 Sep	0.016 ± 0.008	0.016 ± 0.006
		2006	2 Jun	0.041 ± 0.008	0.015 ± 0.004
			4 Jul	0.022 ± 0.005	0.017 ± 0.004
			5 Sep	0.004 ± 0.003	0.007 ± 0.003
	C11	2005	27 Jun	0.12 ± 0.02	0.034 ± 0.008
			12 Jul	0.06 ± 0.01	0.05 ± 0.01
			26 Jul	<0.02	0.035 ± 0.009
			23 Aug	0.09 ± 0.02	0.031 ± 0.009
			14 Sep	0.014 ± 0.008	0.011 ± 0.006
		2006	2 Jun	0.047 ± 0.008	0.015 ± 0.005
			4 Jul	0.024 ± 0.006	0.014 ± 0.004
			5 Sep	<0.006	0.005 ± 0.003
	C12 (Blk.)	2005	27 Jun	<0.02	<0.02
			12 Jul	<0.02	0.011 ± 0.007
			26 Jul	<0.02	0.020 ± 0.007
			23 Aug	0.07 ± 0.02	0.008 ± 0.007
			14 Sep	<0.01	0.008 ± 0.006
		2006	2 Jun	0.031 ± 0.006	0.014 ± 0.005
4 Jul			0.009 ± 0.004	0.012 ± 0.004	
5 Sep			0.005 ± 0.003	0.010 ± 0.003	
Loam	L10	2005	27 Jun	0.07 ± 0.02	0.06 ± 0.01
			12 Jul	0.07 ± 0.02	0.027 ± 0.008
			26 Jul	<0.02	0.018 ± 0.007
			23 Aug	0.09 ± 0.02	0.029 ± 0.008
			14 Sep	0.016 ± 0.008	0.032 ± 0.007
		2006	2 Jun	0.041 ± 0.008	0.018 ± 0.005
			4 Jul	0.028 ± 0.006	0.019 ± 0.005
			5 Sep	0.012 ± 0.003	0.018 ± 0.004
	L11	2005	27 Jun	0.11 ± 0.02	0.05 ± 0.01
			26 Jul	<0.02	0.024 ± 0.007
			23 Aug	0.08 ± 0.02	0.035 ± 0.009
		2006	2 Jun	0.034 ± 0.006	0.017 ± 0.004
			4 Jul	0.020 ± 0.005	0.013 ± 0.004
			5 Sep	0.010 ± 0.003	0.016 ± 0.004
	Lysimeter Lb	2005	27 Jun	0.05 ± 0.02	0.07 ± 0.01
		2006	2 Jun	0.049 ± 0.009	0.010 ± 0.004
			4 Jul	0.038 ± 0.008	0.006 ± 0.004
			5 Sep	0.010 ± 0.004	<0.004
	L12 (Blk.)	2005	27 Jun	<0.02	<0.01
			12 Jul	<0.02	<0.01

Table A10 Measured concentrations of tritium in grass

Soil	Container	Date	Tritium concentration (Bq g ⁻¹)				
			Aqueous	OBT			
Peat	P10	2006	26 Jul	<0.02	<0.008		
			23 Aug	0.10 ± 0.02	0.011 ± 0.007		
			14 Sep	<0.01	0.05 ± 0.01		
			2 Jun	0.035 ± 0.007	0.014 ± 0.005		
			4 Jul	0.009 ± 0.004	0.006 ± 0.004		
			5 Sep	0.004 ± 0.003	0.013 ± 0.004		
		2005	27 Jun	0.06 ± 0.02	0.05 ± 0.02		
			12 Jul	0.04 ± 0.01	0.07 ± 0.01		
			26 Jul	<0.02	0.05 ± 0.01		
			23 Aug	0.10 ± 0.02	0.016 ± 0.007		
			14 Sep	0.011 ± 0.009	0.035 ± 0.010		
			2006	2 Jun	0.040 ± 0.008	0.004 ± 0.005	
				4 Jul	0.007 ± 0.004	0.008 ± 0.004	
				5 Sep	0.005 ± 0.003	0.014 ± 0.004	
				2005	27 Jun	0.05 ± 0.02	0.05 ± 0.02
12 Jul	0.06 ± 0.01				0.06 ± 0.01		
26 Jul	<0.02				0.05 ± 0.01		
23 Aug	0.08 ± 0.02		0.06 ± 0.01				
14 Sep	<0.02		0.017 ± 0.008				
2006	2 Jun		0.028 ± 0.006		0.008 ± 0.005		
	4 Jul		0.007 ± 0.004	<0.005			
	5 Sep	0.002 ± 0.003	0.007 ± 0.003				
	Lysimeter Pb	2005	27 Jun	0.01 ± 0.01	0.04 ± 0.01		
		2006	2 Jun	0.046 ± 0.008	0.005 ± 0.004		
			4 Jul	0.038 ± 0.008	0.006 ± 0.003		
5 Sep			0.010 ± 0.004	0.005 ± 0.003			
P12 (Blk.)		2005	27 Jun	<0.02	<0.02		
			12 Jul	0.008 ± 0.008	0.032 ± 0.008		
	26 Jul		<0.02	0.025 ± 0.008			
	23 Aug		0.08 ± 0.02	0.007 ± 0.006			
	14 Sep		<0.02	0.018 ± 0.008			
	2006		2 Jun	0.025 ± 0.006	0.006 ± 0.005		
		4 Jul	0.005 ± 0.004	<0.005			
		5 Sep	0.006 ± 0.003	0.010 ± 0.003			
		Sandy	S10	2005	27 Jun	0.06 ± 0.02	0.06 ± 0.01
					12 Jul	0.07 ± 0.01	0.06 ± 0.01
					26 Jul	<0.02	0.026 ± 0.007
	23 Aug				0.11 ± 0.02	0.034 ± 0.008	
14 Sep	<0.02				0.04 ± 0.01		
2006	2 Jun				0.044 ± 0.008	0.011 ± 0.004	
	4 Jul			0.036 ± 0.007	0.037 ± 0.007		
	5 Sep			0.013 ± 0.004	0.020 ± 0.004		
	2005			27 Jun	0.10 ± 0.02	0.07 ± 0.02	
				12 Jul	0.06 ± 0.01	0.041 ± 0.009	
				26 Jul	<0.02	0.026 ± 0.007	
23 Aug				0.10 ± 0.02	0.022 ± 0.007		

Table A10 Measured concentrations of tritium in grass

Soil	Container	Date	Tritium concentration (Bq g ⁻¹)		
			Aqueous	OBT	
		14 Sep	0.011 ± 0.010	0.037 ± 0.009	
	2006	2 Jun	0.041 ± 0.008	0.007 ± 0.004	
		4 Jul	0.028 ± 0.006	0.016 ± 0.004	
		5 Sep	0.006 ± 0.004	0.010 ± 0.004	
	Lysimeter Sb	2005	27 Jun	0.02 ± 0.01	0.05 ± 0.01
		2006	2 Jun	0.047 ± 0.008	0.008 ± 0.004
			4 Jul	0.021 ± 0.004	0.013 ± 0.004
			5 Sep	0.008 ± 0.004	0.017 ± 0.004
	S12 (Blk.)	2005	27 Jun	0.02 ± 0.01	0.009 ± 0.01
			12 Jul	<0.02	<0.01
			26 Jul	<0.02	0.014 ± 0.007
			23 Aug	0.09 ± 0.02	0.011 ± 0.006
			14 Sep	<0.01	0.009 ± 0.005
		2006	2 Jun	0.033 ± 0.007	<0.006
			4 Jul	0.017 ± 0.005	0.015 ± 0.004
			5 Sep	0.006 ± 0.003	0.006 ± 0.002

APPENDIX B Comparison with values in predictive models

Concentration factors for tritium used in the calculation of Generalised Derived Constraints (GDC)s: comparison with experimental data from the lysimeter study

B1 APPROACH ADOPTED

From the experimental work described in the main report, the aggregated transfer quotient for the transfer of tritium from sludge amended land to vegetables was estimated to be $2 \cdot 10^{-4}$ Bq kg⁻¹ per Bq m⁻². The tritium was applied in the form of pellets and was almost all in the form of OBT. The transfer quotient was based on data for the second year of growth, ie 1 year after amendment. This was based on the requirement from the Safe Sludge Matrix that crops could only be harvested at least 10 months after the sludge had been applied.

The model used to calculate concentrations in animal products in the estimation of a GDC for sewage sludge was based on the TRIF model (Higgins et al, 1996). This model assumes that tritium was applied to the soil in the form of HTO and HT and not OBT as was the case in the present study.

The results from the experimental study indicated that most of the activity in grass was the result of soil splash. The comparison carried out here relates to uptake via roots, and so it has been assumed that the aggregated transfer quotient derived for vegetables can also be applied to grass. The same quotient has been derived from the methodology used to estimate GDCs. The two sets of values are compared in Table 1.

The estimation of GDCs was based on a delay of 21 days between application of sludge and the commencement of grazing, since this is the shortest interval that is permissible under the Safe Sludge Matrix. A peak value is given in Table 1 together with the average over the period between 21 and 800 days after deposition. The quotient derived for grass that was planted 1 year after deposition is also given. This value can then be compared directly with the experimental data.

Table B1 Comparison of predicted and observed values for the aggregated transfer quotient

	TRIF (HTO) (assume 72% HTO) (Bq kg ⁻¹ per Bq m ⁻²)	Experimental data (Bq kg ⁻¹ per Bq m ⁻²)
Start of first year growing	$1.2 \cdot 10^{-2}$ (Peak)	$4 \cdot 10^{-4}$
Average between 21-800 d Used for GDCs/GDLs	$6.1 \cdot 10^{-4}$	
Start of second growing season	$1.7 \cdot 10^{-7}$	$2 \cdot 10^{-4}$

The main reasons for the differences between the two sets of values are as follows.

- a The TRIF model is for HTO and long term losses are assumed to be much faster than is the case in the experimental study, where most of the activity is in the form of OBT.
- b The model predicted that initial transfer would be higher than was actually observed, because HTO is readily available for plant uptake.

As noted in the main report, the amount of OBT in vegetables is small compared with HTO and this has not been considered here.

B2 CONCLUSIONS

- a The experimental study of the transfer from sewage pellets to plants related to OBT. The data cannot therefore be compared directly with the TRIF model used in the estimation of GDCs and GDLs since this assumes that tritium was in the form of HTO and HT. However if tritium in sewage sludge pellets was assumed to behave the same as HTO then for the first 800 days after application the model predictions would be cautious, but only by a factor of less than 2.
- b For tritium originally applied to the soil in the form of OBT, the long term transfer to vegetables should be modelled based on the approach described in the experimental study, since the aggregated transfer coefficient is much greater than that derived from the TRIF model. However, the comments in the main report concerning the use of the actual values derived in the experimental study must be noted.

B3 REFERENCE

Higgins NA, Shaw PV, Haywood SM and Jones JA. (1996). *TRIF A dynamic model for predicting the transfer of tritium through the terrestrial foodchain*. Chilton, NRPB-R278