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Version 1

Tritium in Scottish Landfill Sites

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Tritium in Scottish Landfill Sites

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Tritium Sources in Landfill Sites in Scotland				
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1 Introduction

This report presents an analysis of the possible sources of tritium that has been detected in samples of leachate from a number of Scottish landfill sites. The report has been prepared by Galson Sciences Ltd (GSL) on behalf of the Scottish Environment Protection Agency (SEPA).

SEPA's duties and responsibilities were established by the Environment Act of 1995. SEPA's primary responsibility is the implementation of a wide range of environmental protection legislation within Scotland. SEPA's remit includes regulation of the use and disposal of radioactive substances.

Twelve landfill sites in Scotland are approved special precautions burials sites that are authorised to receive solid wastes containing very low levels of radioactivity. As part of its responsibilities, SEPA monitors for radioactivity in leachates from these sites. Annual samples have been collected and analysed by SEPA since 1996[1]. Concentrations of tritium in some leachate samples have been found to be greater than concentrations that occur in rainwater or natural groundwater, although the levels of radioactivity detected in leachates are still of low radiological significance. SEPA commissioned GSL to identify possible sources of tritium in landfills and to assess how these sources could contribute to elevated tritium levels in landfill leachates.

Section 2 of this report presents a summary of previous studies on tritium in landfill leachate in the UK, and Section 3 describes potential sources of tritium in domestic and commercial wastes. Section 4 presents a summary of annual monitoring results at Scottish landfill sites, and includes additional information on hydrogeology, tritium levels, and authorised disposals of tritium at the Ness landfill site in Aberdeen. Section 5 synthesises the available information and draws conclusions on the probable source of tritium in landfill leachates. Section 5 includes the results of scoping calculations based on the data obtained from the Ness site. Section 6 summarises the conclusions from the project and presents a number of recommendations for further action by SEPA.

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[1] Her Majesty's Industrial Pollution Inspectorate monitored for radioactivity in landfill leachates prior to 1996.

2 Background

The issue of unexpected tritium concentrations in landfill leachates has been the subject of previous studies. Robinson and Gronow (1996) reviewed leachate compositions from 30 UK landfills that have received mainly domestic wastes. Their study compared tritium levels in landfill leachates and condensates with concentrations of tritium expected in rainfall.

Tritium levels in UK rainfall show an annual cycle, with tritium concentrations in summer rainfall generally about four times greater than concentrations in winter rainfall. Tritium concentrations in rainfall have been declining cyclically since peak concentrations of a few hundred Bq/l were recorded in the UK in 1963 following atmospheric testing of nuclear weapons in the previous decade. Robinson and Gronow (1996) reported that, in 1990, tritium concentrations in rainfall had fallen to about 50 tritium units (TU)[2] (6 Bq/l) in summer and 20 TU (2.4 Bq/l) in winter. The natural action of cosmic rays contributes a tritium content to rainfall of 3 to 10 TU (0.4 to 1.2 Bq/l).

Robinson and Gronow (1996) determined that drinking water tritium concentrations in excess of 10^5 TU (about 10^4 Bq/l) would be needed before acceptable levels of radioactivity were exceeded. The World Health Organization guidelines for drinking water quality do not specify any limit for tritium, but the National Radiological Protection Board (1991) suggests a tritium exposure limit of 10^7 Bq per year for members of the public. On this basis and assuming a drinking rate of 584 l/year, a tritium concentration of up to 1.4×10^5 TU (1.7×10^4 Bq/l) would be acceptable in drinking water (Robinson and Gronow, 1996).

Almost all landfill leachate and condensate samples considered by Robinson and Gronow (1996) contained much higher tritium concentrations than can be accounted for by existing or historic levels in rainfall. The mean concentration for leachate samples for the 30 landfill sites studied was 7,714 TU (926 Bq/l), with a maximum of 39,270 TU (4,712 Bq/l). Tritium concentrations in leachates from sites that had received some industrial wastes were on average about 50 % higher than in leachates from landfills that had received only domestic and commercial wastes. Robinson and Gronow (1996) concluded that some widespread component of UK domestic or commercial waste streams, such as gaseous tritium light sources (GTLSs), is responsible for the concentrations of tritium found in leachates at most sites.

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[2] A tritium unit is equivalent to 1 tritium atom per 10^{18} hydrogen atoms and the activity corresponding to 1 TU is 0.12 Becquerels per litre of water.

3 Tritium Sources

3.1 Introduction

There are several potential sources of tritium in waste that could be disposed of in landfill sites. Kisalu *et al*. (1991) discussed uses of tritium in consumer and industrial items and identified the following devices that contain tritium:

- Gaseous tritium light devices (GTLDs) (up to 1 TBq/device); GTLDs are assembled from a number of GTLSs, with each GTLS containing up to 80 GBq of activity.
- Watches and clocks containing GTLSs (up to 7.5 GBq/device).
- Watches and clocks including luminous paints (up to 0.28 GBq/device).
- Compasses (up to 0.3 GBq)
- Electron tubes (up to 0.0008 GBq)

In addition, Robinson and Gronow (1996) noted that the ‘trimphone’ (produced by British Telecom from 1964 until 1976) had a luminous dial (a GTLS) that contained about 25 mCi (0.9 GBq) of tritium. British Telecom has aimed to recover as many as possible of the trimphones distributed in the UK, which it sends to AEA Technology for storage pending recovery of the tritium. However, it is likely that many trimphones could have been disposed of in domestic landfill sites at the end of their use up to the mid-1980s.

Mobbs *et al*. (1998) reported that the Ministry of Defence (MoD) uses a large number GTLSs in devices such as route markers and night sites, although most of these devices have activity of less than 20 GBq.

Currently, a redundant GTLD comprising several GTLSs presents the largest single potential source of tritium in landfill sites. The following sections discuss the regulatory framework concerning GTLDs, the number of GTLDs and GTLSs distributed in the UK, and the characteristics of redundant devices.

3.2 Regulations Concerning the Use and Disposal of GTLDs

The main pieces of legislation concerning GTLDs in the UK are the Radioactive Substances Act 1993, the Ionising Radiations Regulations 1985 and, where the GTLD is used as a fire safety sign, the Health and Safety (Safety Signs and Signals) Regulations 1996. The application of these regulations, and a series of relevant standards, to the use of GTLDs has been described in detail by Mobbs *et al.* (1998).

Of particular significance to the control of GTLDs is the Radioactive Substances (Gaseous Tritium Light Devices) Exemption Order 1985. The exemption order is applicable to three classes of device:

Class A A GTLD in which the activity does not exceed 20 GBq.

Class B A GTLD in which the activity does not exceed 1 TBq, and the activity of each sealed container (GTLS) in the GTLD does not exceed 80 GBq.

Class C A GTLD installed, or awaiting installation, in a vessel or aircraft, or a vehicle or equipment used, or intended for use, by the armed forces of the Crown.

Under the exemption order, all persons are exempted from registration under the Act in respect of keeping and use of GTLDs, subject to certain conditions. Conditions of significance to the present study are the requirements that:

- The activity in the form of tritium oxide and other water soluble compounds of tritium in any sealed container does not exceed 2% of the total activity, or does not exceed 100 MBq if the total activity is less than 5 GBq.

- Class B and Class C GTLDs should bear a statement of the manner in which the article can be disposed of as waste without specific authorisation.

Under the exemption order, radioactive waste consisting of GTLDs does not require authorisation for disposal subject to conditions, including:

- A GTLD containing not more than 20 GBq of activity may be disposed of as normal refuse provided it is dispersed in refuse which is non-radioactive waste and not more than one such GTLD is disposed of in any 0.1 m^3 of non-radioactive waste. Such GTLDs must not be disposed of in any part of a site that is used for the deposit of radioactive waste only.

- A GTLD with an activity greater than 20 GBq must be disposed of by a person authorised to dispose of such radioactive waste or by a GTLD manufacturer.

- Records must be kept at the premises (where the GTLD has been used) showing the nature and activity of disposal and the means and date of disposal.

3.3 The Use of GTLDs and GTLSs

The main use for GTLDs in the UK is as “exit” signs and similar signs where electrical wiring and maintenance is to be minimised. The light source is provided by a number of component GTLSs. Each GTLS is manufactured by sealing gaseous tritium in a glass capsule, the inside surface of which has been coated with a phosphor. Beta particles produced by the radioactive decay of tritium to ^{3}He excite the phosphor causing it to emit light. The intensity of the light diminishes as the tritium decays. The useful life of a GTLS is typically 10-12 years (tritium has a half-life of 12.3 years).

As part of the current survey, the distributors of GTLDs in the UK were contacted in order to determine the volume of such devices currently distributed in the UK and their intended disposal routes. SRB Technologies (previously Saunders-Roe Ltd), based in Datchet, Berkshire, is the main distributor of GTLDs (Betalights) in the UK. Saunders Roe absorbed the other main distributor (Brandhurst) in 1984. Purpose-built signs are manufactured in Canada for distribution by SRB in the UK. All signs carry a radioactivity symbol and instructions explaining that the signs should be returned to the distributor when redundant. SRB returns redundant Betalights to Canada.

SRB’s rate of distribution of Betalights to commercial organisations in the UK was at its maximum about 10 years ago when, in one year, about 1,000 Betalights were sold. For a period of about 5 years around this time of peak production, around 150 to 200 Betalights were distributed each year. However, the availability of cheaper alternatives (such as photoluminescent materials or battery powered signs) has meant that there is no longer a demand for Betalights and none are distributed. Currently, about 100 redundant signs are returned by commercial organisations to SRB each year for return to Canada. SRB considers that most of the signs it distributes in the UK are returned, although no records were available to this project to verify this. Other redundant GTLDs are sent to a recycling service run by Safeguard International.

One organisation, Surelite (based in Corby, Northamptonshire), does manufacture GTLDs in the UK, but the vast majority of these GTLDs are exported to the US. However, redundant GTLDs are returned to Surelight in the UK, where they are currently stored.

Mobbs *et al.* (1998) presented a general review of the use of GTLSs and noted that, at the peak of production, the tritium in GTLD exit signs amounted to less than 9% of the average annual total production of GTLSs in the UK. Between 1980 and 1995, an average of about 125,000 GTLSs with an activity of less than 20 GBq, and 21,000 GTLSs with an activity of greater than 20 GBq, were produced annually. The MoD uses the majority of these GTLSs, in many cases, in activities abroad. Mobbs *et al.*

(1998) reported that some 60% of GTLSs supplied to the UK are exported in assembled articles. The MoD stores redundant devices greater than 20 GBq pending tritium extraction, and disposes of devices of lower activity with normal refuse unless extraction of the GTLSs is difficult or the devices are too large to be disposed of in this way.

The possibility of GTLDs of activity greater than 20 GBq being disposed of in landfill sites, in contravention of the GTLD exemption order, cannot be discounted, although it has not been possible to confirm that GTLDs have been disposed of in UK landfills. Nevertheless, the rate of disposal of GTLDs in landfill sites could currently be close to a maximum because the many Betalights distributed ten years ago are coming to the end of their period of use. Thus, the concentrations of tritium found in landfill leachate could increase considerably in the near future, to the extent that GTLDs are disposed of in landfills. However, provided the demand for GTLDs does not return, and that these devices are the major source of tritium in UK landfills, the concentrations of tritium in leachates should eventually begin to diminish. Alternatively, GTLDs of activity less than 20 GBq, disposed of in domestic waste under the GTLD exemption order, could be responsible for elevated tritium levels in landfill leachate.

3.4 Characteristics of GTLDs

In order to understand the possible effects of disposal of redundant GTLDs in landfills, it is necessary to consider the amounts and forms of tritium likely to be contained in GTLDs. Harding (1993) reported the results of two sets of analyses that have been undertaken to characterise GTLDs. First, Harding (1993) summarised the findings of an analysis of redundant GTLDs that had been undertaken by AEA Technology in 1989. Analysis of GTLDs that were over 10 years old found that only a small percentage of the tritium present existed in gaseous form. Devices that were 10 years old contained about 10% gaseous tritium, and devices older than 15 years contained on average less than 2% gaseous tritium. Based on this analysis, it was concluded that gaseous tritium slowly exchanges with combined hydrogen atoms in the component materials to form a range of tritiated species, water being the predominant form. Thus, at some time after manufacture, GTLDs will fail to satisfy the exemption order condition that the activity in the form of tritium oxide and other water-soluble compounds of tritium in any sealed container does not exceed 2% of the total activity.

These findings prompted Her Majesty's Inspectorate of Pollution to commission further analyses of GTLDs (Harding, 1993). A number of GTLDs, between 2 and 14 years old, were acquired from two manufacturers. The GTLSs were extracted from assemblies containing between 11 and 16 GTLSs that had been used in "exit" signs. Fourteen-year old GTLSs from one manufacturer were found to contain about 18 GBq of activity; four-year old GTLSs were found to contain about 36 GBq of activity. The manufacturers' labelling indicated that the GTLSs contained initial activities of between about 45 and 75 GBq, but Harding (1993) found that the GTLSs would have contained an initial tritium inventory of either about 39 GBq or about 13 GBq, depending on the manufacturer. The manufacturers' labels may represent maximum possible amounts of tritium contained in each GTLS.

Harding (1993) confirmed that the percentage of tritium present as a gas in a GTLS decreases with time. About 80% of the tritium was present in a gaseous form in two-year old devices and about 40% of the tritium was present in a gaseous form in items that were 14 years old. Up to 1% of this gaseous tritium was found to be in the form of water vapour. The total amount of tritium found to have been adsorbed by the Pryex glass tubing ranged from about 3 GBq to about 15 GBq, although the amount adsorbed did not appear to show any clear dependence on the age of the GTLS.

As part of the analysis, Harding (1993) undertook experiments to determine the rate at which tritium adsorbed in the Pyrex glass tubing of the GTLSs could be released into water at ambient temperature. Several GTLSs were crushed under water and the amounts of tritium in the water were monitored. In one test on five-year old GTLSs, about 90% of the tritium contained in each GTLS was released in 21 days. This amounted to the release of about 6 GBq of tritium per GTLS. Harding (1993) determined through further analysis that the tritium was likely to be present in the GTLS materials as T_2O.

The results presented by Harding (1993) have important implications on the levels of tritium that could occur in leachate if a single GTLD was disposed of at a landfill site. One redundant "exit" sign comprising 16 GTLSs could contain on the order of 100 GBq of tritium adsorbed in the materials of the GTLD. If the GTLSs were broken, the adsorbed tritium could be removed rapidly from the GTLSs and enter the leachate primarily as T_2O.

4 Survey of Scottish Landfill Sites

Twelve landfill sites in Scotland are authorised by SEPA for the disposal of solid wastes containing very low levels of radioactivity. The leachate from these sites is monitored for radioactivity. The National Radiological Protection Board (NRPB) undertakes this monitoring on behalf of SEPA, and collects and analyses one sample of leachate from each site on an annual basis.

As part of this project, the NRPB was contacted and asked to provide information on levels of tritium found in leachate samples taken from the 12 Scottish sites over a number of years. The site locations and sampling points are described in Table 1 and the measured concentrations of tritium in the leachate from each site between 1995 and 1999 are shown in Table 2. It is apparent from Table 2 that tritium levels above expected background levels (a few Bq/l) have been recorded in all but one of the landfill sites.

In 1999, on behalf of SEPA, the NRPB undertook more comprehensive sampling and analysis of leachate from the Ness Tip. Ness is one of the landfills showing elevated levels of tritium in leachates, and is the only Scottish site where more detailed monitoring information is available. Therefore, the remainder of this section focuses on the conditions (Section 4.1) and disposals of radioactive waste (Section 4.2) at the Ness Tip.

4.1 The Ness Landfill Site

The Ness Tip was visited during the course of this project in order to gain a clear understanding of the layout of the site and the likely directions of leachate migration through the landfill and underlying units. Aberdeen City Council is currently responsible for operating the site, and a representative of the council led the site visit and provided information on site hydrogeology.

Designated areas for infill at Ness and the neighbouring Tullos Hill sites are shown on Figure 1. The two sites are about 400-500 m apart, separated by a conservation area. Domestic and industrial wastes were disposed of at the Tullos Hill site between 1970 and 1983 and have been disposed of at the Ness Farm Landfill site since 1983. The volume of industrial wastes disposed of at the site is about twice the volume of domestic wastes. Disposal of industrial wastes ended in 1999 and disposal of domestic wastes is expected to end this year when the Ness site is full.

Table 1. Leachate sampling locations at landfill sites in Scotland.

Area	**Location**	**Site Status**	**Sample Location**
Aberdeen City	Ness Tip	Working	Purpose-built pool
City of Edinburgh	Braehead	Closed	Stagnant pool at lowest point of site
City of Glasgow	Summerston Tip	Working	Purpose-built pool
Clackmannanshire	Black Devon	Working	Pipe that feeds leachate to a river
Dundee City	Riverside	Working	The Tay Estuary beside the site
East Dunbartonshire	Birdston Tip	Closed	Shallow burn at edge of site
Fife	Balbarton	Closed in 1999	Purpose built pool
Fife	Melville Wood	Working	Leachate treatment area
Highland	Longman Tip	Working	Pond at edge of site before 1997; purpose-built system since 1997
North Lanarkshire	Dalmacoulter	Closed in 2000	60-ft-deep borehole
North Lanarkshire	Kilgarth	Working	Pipe feeding leachate to a pond before 1997; culvert nearby since 1997
Stirling	Lower Polmaise	Working	Pumped from a sealed cell

Table 2. Tritium concentrations (Bq/l) in leachate from landfill sites in Scotland between 1995 and 1999.

Area	**Location**	**1995**	**1996**	**1997**	**1998**	**1999**
Aberdeen City	Ness Tip	149.4 ± 6.4	293 ± 22	<25	161 ± 22	136 ± 19
City of Edinburgh	Braehead	<25	69 ± 14	<25	<25	<25
City of Glasgow	Summerston Tip	1381 ± 17	885 ± 50	641 ± 43	252 ± 26	275 ± 25
Clackmannanshire	Black Devon	<25	<25	<25	<25	<25
Dundee City	Riverside	<25	<25	58 ± 18	<25	<25
East Dunbartonshire	Birdston Tip	<25	<25	<25	38 ± 18	<25
Fife	Balbarton	65.4 ± 5.1	40 ± 13	36 ± 18	64 ± 18	79 ± 17
Fife	Melville Wood	<25	<25	55 ± 19	48 ± 19	77 ± 17
Highland	Longman Tip	<25	<25	<25	36 ± 19	<25
North Lanarkshire	Dalmacoulter	399.2 ± 9.4	629 ± 37	170 ± 22	197 ± 24	71 ± 17
North Lanarkshire	Kilgarth	185.8 ± 7.0	<25	<25	<25	<25

Stirling	Lower Polmaise	208.5 ± 7.2	<25	196 ± 23	<25	114 ± 19

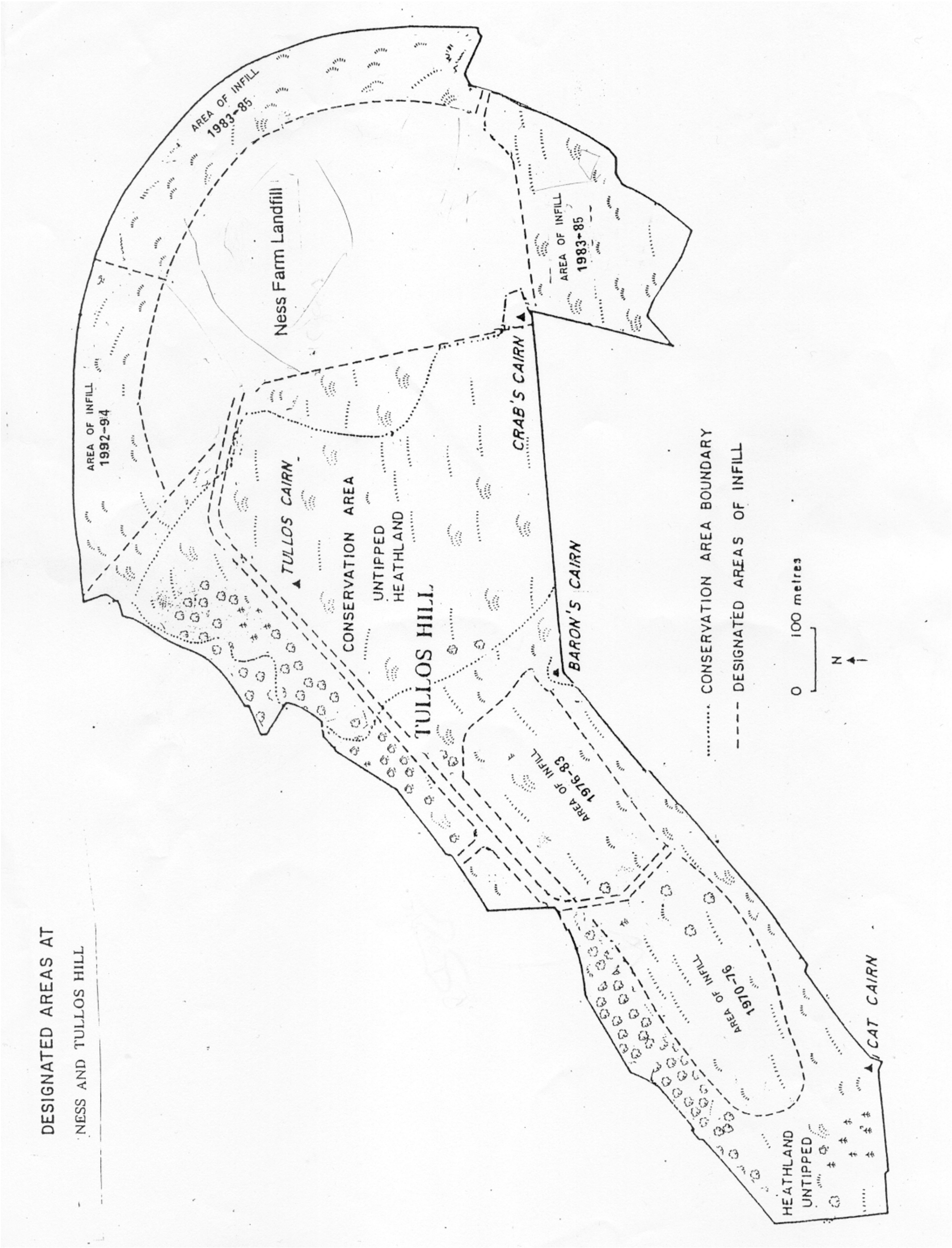

DESIGNATED AREAS AT
NESS AND TULLOS HILL
AREA OF INFILL 1983-85
Ness Farm Landfill
AREA OF INFILL 1983-85
AREA OF INFILL 1992-94
CRAB'S CAIRN
TULLOS CAIRN
CONSERVATION AREA
UNTIPPED HEATHLAND
TULLOS HILL
BARON'S CAIRN
AREA OF INFILL 1976-83
AREA OF INFILL 1970-76
CAT CAIRN
HEATHLAND UNTIPPED
CONSERVATION AREA BOUNDARY
DESIGNATED AREAS OF INFILL
0
100 metres
N

Figure 1. Designated areas for infill at the Ness and Tullos Hill sites.

Site topography and water and gas monitoring and sampling points are shown on Figure 2. The Ness site is bounded to the north and east by the main railway line between Aberdeen and Edinburgh (which lies at about 35 m O.D. to the east and drops to about 20 m O.D. to the north). Crab's Cairn (at about 70 m O.D.) marks a high point at the southwest corner of the Ness site. The ridge between Baron's Cairn (about 80 m O.D.) and Cat Cairn (about 85 m O.D.) bounds the southeast edge of the Tullos Hill site.

At the Ness site, precipitation and surface runoff is likely to infiltrate the landfill and flow down through the waste to saturated zones. Grampian Soil Surveys (1996) undertook a hydrogeological assessment of the Ness Tip and concluded that, in the northern part of the site, groundwater generally flows in a northeasterly direction from the higher ground underlying the landfill. In the southern part of the site, groundwater movement is generally towards the east. Groundwater discharges along the cliff face to the east of the site.

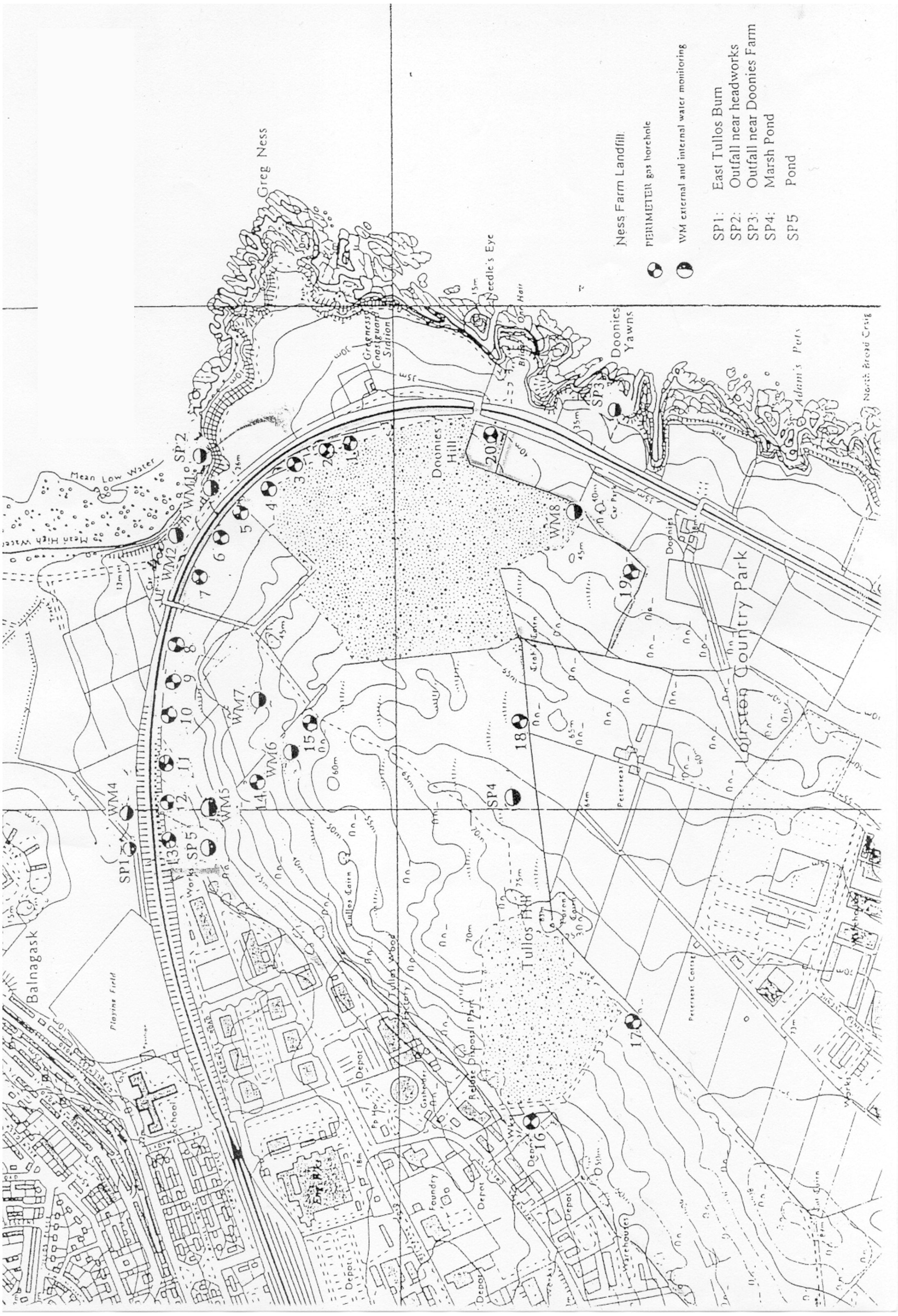
The NRPB took leachate samples on two occasions in 1999 from 8 locations around the site. The results of the tritium analysis are reproduced in Table 3. All previous leachate samples from the Ness Tip were extracted from SP5 (see Tables 1 and 2).

As shown in Table 3, high concentrations of tritium occur in the groundwater at the sampling points and water monitoring points around the cliff face (SP2, WM2, WM1, and SP3). The presence of leachate at these locations is consistent with easterly to northeasterly groundwater flow from the site through permeable units.

The levels of tritium in the leachate at WM6 were below detection limits. This monitoring point is in the conservation area (see Figure 1) hydrologically upstream from the Ness landfill and, thus, tritium would not be expected to occur there.

Table 3. Tritium concentrations (Bq/l) in leachate taken from different sampling locations at the Ness Tip on 19/2/99 and 24/3/99.

Sample Location	**19/2/99**	**24/3/99**
SP5 (Pond)	<25	124
SP1	94	-
SP2 (Outfall)	573	1447
SP3 (Outfall)	3800	-
WM2	-	3260
WM1	-	1152
WM6	-	<25
WM8	-	12040


Ness Farm Landfill
PERIMETER gas borehole
WM external and internal water monitoring
SP1: East Tullos Burn
SP2: Outfall near headworks
SP3: Outfall near Doonies Farm
SP4: Marsh Pond
SP5 Pond
Greg Ness
Needle's Eye
Doonies Yawns
Greg Ness Coastguard Station
Doonies Hill
Tullos Hill
Loirston Country Park
Balnagask
Mean Low Water
Mean High Water
Tullos Wood
School
Foundry
Depot
Works
Playing Field
Refuse Disposal Plant
Car Park
Doonies
Adam's Pots
North Broad Craig
SP1
SP2
SP3
SP4
SP5
WM1
WM2
WM4
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WM7
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Figure 2. Topography and monitoring and sampling points at the Ness landfill site.

The Ness Tip was originally designed on the basis that leachate would be diluted and dispersed in groundwater flowing through the site. However, in 1995 and 1996 shallow drains (a few metres deep) were installed along boundaries to the north of the site and to the south and east of the site to prevent leachate reaching surface water in two public access areas outside the site boundaries. The drain to the north (running from near borehole 8 to near borehole 13) discharges to the pond (SP5). Tritium concentrations at the pond and to the north of the site (SP1) are an order of magnitude less than occur in the leachate along the cliff faces. Periodically water is removed from the pond by tanker and is recirculated at the site, for example, through spaying for dust control. Typically, the pond collects between 10,000 and 20,000 litres of water per day.

The drains to the south and east of the site (running along the site boundary from near borehole 20 to west of borehole 19) discharge to a covered sump (WM8). Periodically the leachate from this sump is pumped upwards through an underground pipe and discharged at a point in the landfill just below Crab's Cairn, in an area where landfilling has been completed (Figure 1). The amount of leachate recirculated in this way is unknown. The tritium concentration in the sump (WM8) is much higher than at other sampling locations, and is close to the maximum level that would be accepted in drinking water. It is apparent that a significant tritium source exists in the catchment area for the sump. Recirculation of the leachate collected in the sump could be gradually increasing concentrations of tritium in this area above values detected at other locations. The elevated tritium concentrations detected in leachate at the SP3 outfall indicate that some leachate is migrating from the site in an easterly direction below the WM8 drainage system. Potentially, the tritium concentration in the outfall could increase if the source of this tritium is affected by leachate recirculation.

4.2 Authorised Disposals of Radioactive Waste at the Ness Tip

The Ness Tip is authorised for the disposal of wastes containing low levels of radioactivity. The quantities of past disposals of tritium wastes at the site were investigated in order to support an assessment of whether such disposals could give rise to the observed levels of tritium in the leachate.

The Aberdeen Royal Infirmary and University are the principal producers of tritium wastes for disposal at the Ness Tip. The Radiation Protection Advisor (RPA) at the University's Department of Biomedical Physics supervises the disposal of all radioactive wastes from the hospital and university. Wastes are disposed of in sealed polythene bags, and hand-written records of the number of bags and types of waste sent for disposal are maintained by the RPA. These disposal records were inspected during the course of this project. Several hospital and university departments generate radioactive wastes, and records of the number of bags of such waste generated by each department have been kept since 1973. Records kept since mid-1994 include measurements of the amounts of the various radioactive isotopes generated by each department. These measurements are made by research students at the university. In a few cases measurements of combined ^{3}H and ^{14}C activity have been recorded, but for this project such measurements have been assumed to relate only to tritium. The RPA reported that tritium wastes are almost all in a solid form and contain no free tritium and negligible tritiated water.

Data on tritium disposals from each department were compiled as part of this project. The total amounts of tritium waste generated annually (between 1995 and 1999) by the hospital and university are shown in Figure 3. On average, about 1 GBq of tritium is disposed of at the site each year.

The Ness Tip supervisor weighs the bags containing radioactive waste on receipt at the site, but waste radioactivity is not checked. Specific areas have been designated for authorised disposals of radioactive waste at the Ness site as shown on Figure 4. Based on consideration of the locations of these designated disposal regions and the expected directions of leachate migration (as discussed in Section 4.1), it appears unlikely that all the tritium present in the landfill leachate can have been generated by authorised disposals of tritium. Indeed, because leachate migration is likely to be easterly to northeasterly in the vicinity of the authorised disposal areas, it is unlikely that any tritium from such areas could have entered the catchment area for the sump at WM8, where the highest levels of tritium have been found. Thus, some other source(s) of tritium must be present in the landfill site.

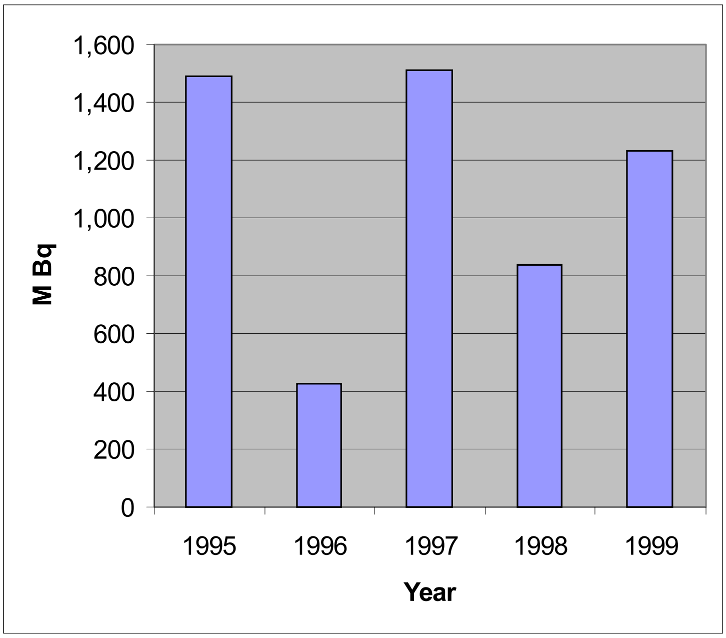

Figure 3. The amount of tritium sent annually to the Ness Tip between 1995 and 1999 for authorised disposal.

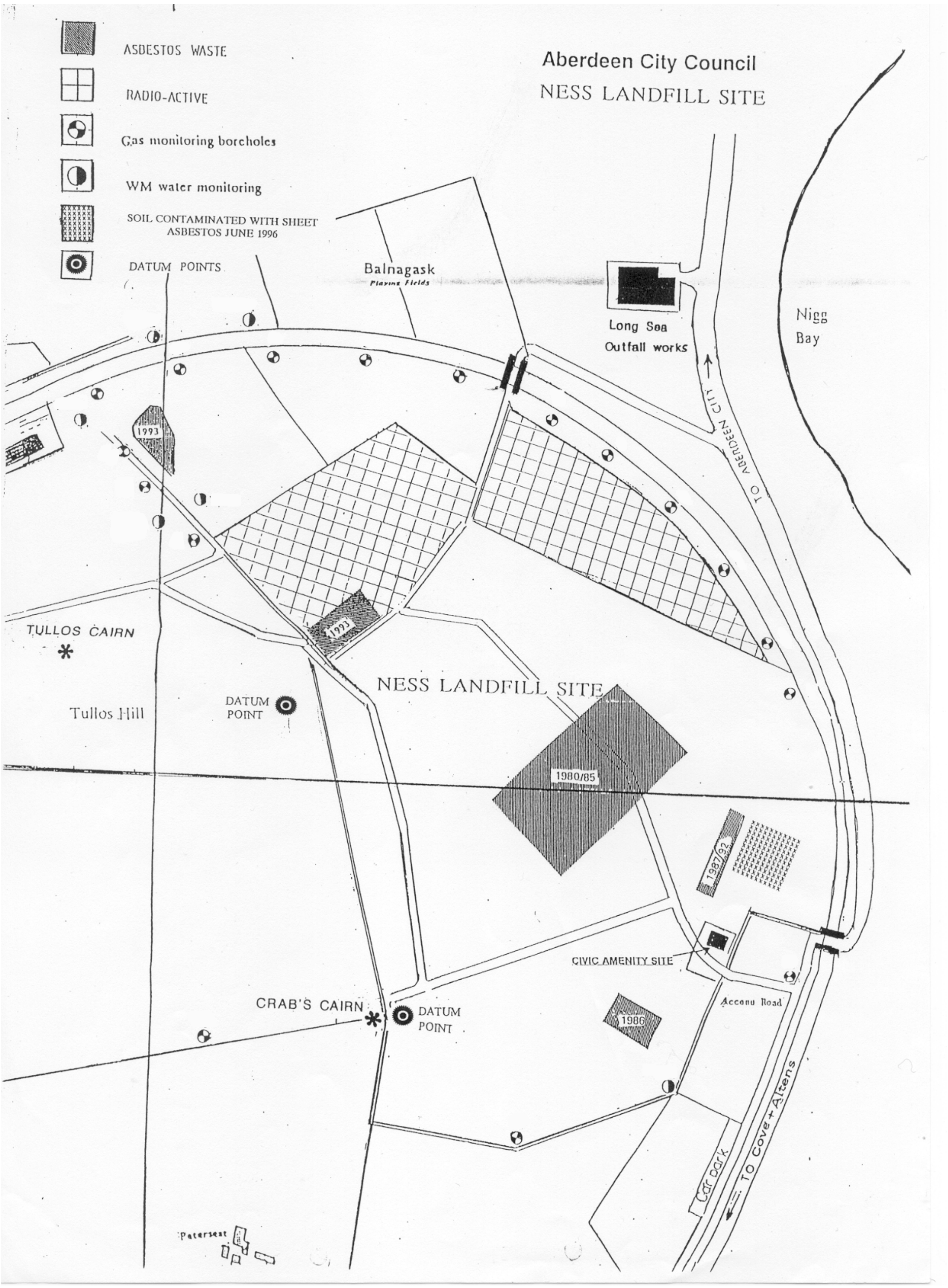
ASBESTOS WASTE
RADIO-ACTIVE
Gas monitoring boreholes
WM water monitoring
SOIL CONTAMINATED WITH SHEET ASBESTOS JUNE 1996
DATUM POINTS
Aberdeen City Council
NESS LANDFILL SITE
Balnagask
Playing Fields
Long Sea Outfall works
Nigg Bay
TO ABERDEEN CITY
1993
1993
TULLOS CAIRN
Tullos Hill
DATUM POINT
NESS LANDFILL SITE
1980/85
1987/92
CIVIC AMENITY SITE
Access Road
CRAB'S CAIRN
DATUM POINT
1986
Car park
TO Cove + Altens
Paterseat

Figure 4. Areas designated for authorised disposals of radioactive waste at the Ness landfill site.

5 Possible Sources of Tritium in Leachate

Several domestic and commercial uses of tritium have been identified (see Section 3), which could represent sources of tritium in landfills, in addition to authorised disposals of tritium wastes. This study has considered the possible impact of the disposal of tritium-containing devices on tritium concentrations in leachate in the context of the Ness landfill site.

Trimphones containing tritium were manufactured until 1976 (Section 3). At present, one trimphone will contain no more than about 0.2 GBq of tritium (assuming tritium decay of two half lives). Tritium concentrations on the order of 10^3 Bq/l have been observed in leachate at the Ness Tip. Assuming a uniform distribution of tritium from one trimphone, such concentrations of tritium could be achieved in $2x10^5$ l of leachate. The Ness site has a surface area of approximately $6x10^5$ m^2. Assuming a hydraulically effective rainfall (HER) of about 0.2 m/year at the site, a total of about $3x10^5$ l/day of leachate could be generated. Thus, one trimphone could generate concentrations of tritium observed at the Ness site for about 0.7 days. To sustain observed levels of tritium concentrations for a year would require in excess of 500 trimphones to be distributed in the landfill. The value of HER used in this analysis is towards the low end of the range of expected values. This represents the worst case because increases in HER would increase leachate production and would tend to reduce tritium concentrations.

As discussed in Section 3, a single redundant GTLD could contain about 100 GBq of tritium adsorbed in the materials of the device in a form that could readily dissolve in leachate should the device be broken in a landfill site (although such disposal would contravene the GTLD exemption order). Again, assuming a uniform distribution of tritium from such a GTLD, a tritium concentration of 10^3 Bq/l could be achieved in 10^8 l of landfill leachate. Assuming leachate is generated at about $3x10^5$ l/day, one broken GTLD could generate observed concentrations of tritium for about 300 days. Several GTLDs disposed of at different times and at different locations around a landfill site could generate observed levels of tritium for many years. Alternatively, legitimate disposal of a few tens of GTLSs of lower activity (less than 20 GBq), for example, by the MoD, could generate similar tritium concentrations if many of these devices were broken at the time of, or after, disposal.

Authorised disposals of tritium at the Ness Tip typically occur on a fortnightly to monthly basis amounting to about 1 GBq per year ($2.7x10^6$ GBq/day) (see Section 4). The part of the Ness site designated for radioactive waste disposal has a surface area of approximately 10^5 m^2. Assuming a hydraulically effective rainfall of about 0.2 m/year at the site, a total of about $5x10^4$ l/day of leachate could be generated in this region. Assuming tritium is extracted by the leachate at a uniform rate after disposal, then average tritium concentrations of about 50 Bq/l could be sustained in this leachate. Thus, the tritium concentration that could be generated by such wastes is at least an order of magnitude less than is observed in leachate at the site.

Individual doses resulting from exposure to tritium in landfills can be estimated for a range of exposure scenarios. For example, inadvertent ingestion of leachate (2.5 l per year) containing the highest measured values of tritium concentration (12,040 Bq/l) would result in an annual dose of 0.0005 mSv. A dose conversion factor of 1.8×10^{-11} Sv/Bq has been used for an adult ingesting tritiated water. It is conceivable, although unlikely, that such exposure could occur during the annual cleaning of the sump (WM8). In the unrealistic event of an individual drinking undiluted leachate (with a tritium concentration of 12,040 Bq/l) at a rate of 584 l/year (see Section 2), the annual dose would be 0.13 mSv.

Mobbs *et al*. (1998) considered other pathways for exposure of workers to tritium during landfill operations involving inhalation of dust while moving waste, inhalation of the gas phase from GTLD breakage, and skin absorption from gas phase release of GTLD breakage. The total individual effective dose for these exposure pathways was calculated to be 0.17 mSv/year. Other scenarios that could affect doses to workers include ingestion while spraying leachate for dust control.

Guidance on requirements for authorisation of disposal facilities on land for low and intermediate level radioactive wastes, published by SEPA, the Environment Agency and the Department of the Environment for Northern Ireland (Environment Agency *et al.* 1996), specifies that the effective dose to a representative member of the critical group from a facility shall not exceed a source-related dose of 0.3 mSv/year.

6 Conclusions and Recommendations

This study has aimed to identify possible sources of tritium in leachate samples taken from a number of landfill sites in Scotland. The landfill sites of concern are the 12 approved special precautions burials sites at which authorised disposals of small amounts of radioactive wastes have taken place. The study has aimed to determine whether authorised radioactive waste disposals could be responsible for the high levels of tritium found in the leachate, or whether some other source of tritium is responsible.

6.1 Conclusions

The study has focused on the Ness Tip because it has been subject to more detailed leachate sampling than other sites. The study has found that authorised disposal of radioactive waste is not wholly responsible for the elevated levels of tritium found in the leachate at the site, based on consideration of:

- the locations for authorised disposals of radioactive wastes at the site;
- the quantities of such radioactive wastes disposed of at the site;
- the directions of water movement at the site; and
- the locations of leachate sampling points.

A previous study by Robinson and Gronow (1996) reported higher than expected levels of tritium in leachate samples taken from many landfill sites in the UK. These high levels of tritium were found at sites where there have been no authorised disposals of radioactive wastes. Consistent with the current study of the Ness site, it may be concluded that tritium sources must be present in domestic or industrial wastes.

The most significant industrial sources of tritium identified under this project are GTLDs used in “exit” signs and similar applications. A high proportion of exit signs has been reported as being returned to the manufacturer, but it is possible that some have been consigned to landfill sites in contravention of the GTLD exemption order. There is evidence that a large proportion of the tritium in old GTLDs is adsorbed in the materials of the device and that, on disposal, this tritium would be readily leachable. Indeed, within a few years of manufacture, GTLDs will fail to satisfy the GTLDs exemption order condition that the activity in the form of tritium oxide and other water-soluble compounds of tritium in any sealed container does not exceed 2% of the total activity.

Experiments on GTLSs crushed under water revealed that about 90% of the tritium contained in the materials of a GTLS could be released in 21 days (Harding, 1993).

The rate of desorption of tritium from a GTLS disposed of in a landfill will depend on the extent to which the device is broken, allowing water to access its internal surfaces, and rate of flow past the device (which will vary seasonally).

The amount of tritium contained in one redundant GTLD is about two orders of magnitude greater than is contained in the radioactive waste from authorised disposals at the Ness site in one year. Several GTLDs disposed of at different times and at different locations around a landfill site could generate levels of tritium observed at Ness for many years. Alternatively, legitimate disposal of a few tens of GTLSs of lower activity (less than 20 GBq) throughout the site could generate such tritium concentrations if many of these devices were broken.

The highest concentrations of tritium at the Ness site have been found in leachate at a sump into which drains discharge. The leachate collected at the sump is re-cycled through the landfill. This leachate re-cycling process is likely to be responsible for elevating tritium concentrations locally within the Ness site. Continued re-cycling of leachate could lead to activity concentrations above acceptable drinking water levels. It is unlikely that tritium concentrations in the leachate at the Ness site will be affected by future disposals of GTLDs because the site ceased to accept industrial wastes in 1999, and disposal of domestic wastes is expected to end this year when the site is full.

6.2 Recommendations

Continuation of the routine, annual monitoring of radioactivity in landfills as currently conducted is of little benefit because it is largely unrelated to either the spatial or temporal variability in hydrological conditions. A more extensive monitoring programme that does account for spatial and temporal variability has been conducted at Ness. This has revealed tritium levels significantly higher than found in the routine annual sample. Instrumentation may be available to detect and analyse high levels of tritium activity on site. If available, such instrumentation could be used to locate areas where more detailed monitoring should be undertaken. This more extensive monitoring will allow SEPA to work with the site operator on developing revised operational or remedial procedures to ensure that tritium does not pose a significant risk to health. Also, a more detailed assessment of the potential exposure of workers and members of the public to tritium in landfill leachate should be undertaken, based on site specific operational and hydrogeological conditions. Such an assessment should include consideration of the effects of seasonal variations in rainfall and leachate generation on tritium concentrations.

Elevated levels of tritium activity have also been found at several other landfill sites in Scotland (Table 2 in Section 4). An examination of the available hydrological information and the location of leachate monitoring points for these sites is recommended, followed by a more detailed sampling and tritium monitoring programme.

SEPA should consider providing guidance to inspectors and operators concerning the potential for raising activity levels through re-cycling of leachate. Guidance could highlight the possibility of on-site monitoring to check that activity in any leachate that is discharged from the site and/or accessible (e.g., in open drains) is below levels of concern.

This study has drawn attention to potential problems concerning implementation of the Radioactive Substance (GTLD) Exemption Order 1985. In particular, within a few years of manufacture, GTLDs will fail to satisfy conditions concerning limits on the amount of tritium contained in a non-gaseous form. Gaseous tritium in GTLDs is gradually absorbed in the component materials of the device to form a range of tritiated species. SEPA should review exemption orders relevant to the accumulation and disposal of radioactive wastes, and consider any necessary revisions to the GTLD Exemption Order.

7 References

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