

Low Level Waste Repository

Environmental Safety Case

Assessment of Uncertainty in the LLWR ^{14}C inventory




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Issue 1

A report prepared for and on behalf of
the LLW Repository Ltd

National Nuclear Laboratory



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EXECUTIVE SUMMARY

The Low Level Waste Repository (LLWR) is undertaking a programme of work leading to the production of an Environmental Safety Case by May 2011 (2011 ESC). The 2011 ESC will be submitted to the Environment Agency (EA) in order to satisfy a Requirement in the LLWR's current authorisation.

As part of work to support a submission to the Environment Agency on 1st May 2008, NNL undertook an assessment of the radiological impact of ^{14}C -labelled gases. This included a review of the main contributors (greater than 1%) to the ^{14}C inventory in terms of the nature of the waste streams with which it is associated and the potential form of the ^{14}C .

In developing the 2011 ESC, the LLWR would like to develop their knowledge of the ^{14}C inventory in more detail and to consider a wider range of waste streams that might be associated with significant ^{14}C . The assessment presented in this report supports an improved and more realistic consideration of the impact of ^{14}C -labelled gas and the release of ^{14}C via the groundwater pathway.

The Schedule 9 Requirement 2 assessment identified a future vaults inventory for ^{14}C of 7 TBq, leading to a dose from the gas pathway of approximately 10^{-7} y^{-1} . Assessments of the confidence in this value and the presence of ^{14}C in gas-generating waste materials were undertaken.

Analysis of the 2007 National Inventory showed a total of 43 TBq ^{14}C activity in LLW: a reduction from the 2004 National Inventory of 68 TBq. Further analysis showed that the ^{14}C activity associated with LLW identified for disposal to the LLWR was reduced from 7 TBq in the 2004 National Inventory to around 1.7 TBq in the 2007 National Inventory. Assuming a linear relationship between activity and dose, this could lead to a reduction in the dose from the gas pathway to around 10^{-8} y^{-1} .

Assessment of the waste streams contributing the majority of this activity has shown that the estimated ^{14}C inventory is likely to be conservative. Data for many waste streams are provided prior to potential treatments, which may remove or reduce the ^{14}C content.

In the process of reviewing the 2007 National Inventory source data with the Sellafield consignor, a number of errors were discovered, which reveal a significant overestimate in the forecast ^{14}C inventory for particular waste streams. Corrected data were supplied by Sellafield Ltd for some waste streams, but investigations are ongoing, and the findings of this report may need to be reviewed once the complete set of revised data is available. The errors in the construction of the radionuclide fingerprints for Sellafield waste streams have been shown to account for around 100 to 200 GBq of ^{14}C activity.

Magnox future decommissioning wastes, a source of ^{14}C activity, are to be reviewed by Magnox, with potentially a large proportion of the volume being reclassified as VLLW, thereby reducing overall activities.

An assessment of waste form has been undertaken. This has shown that the majority of the ^{14}C activity does not reside in cellulosic material, which accounts for between 60 and 800 GBq of ^{14}C . The bulk of the activity is surface contamination. However, the incineration of cellulose and metal melting may reduce the disposed ^{14}C activity.

It is concluded that the future forecast ^{14}C inventory for the LLWR may potentially fall to less than 800 GBq if radionuclide fingerprints are corrected in line with the findings from the discussions with consignors.

The work undertaken for the Requirement 2 submission used a future vaults ^{14}C inventory of 7.35 TBq, coupled with a cellulosic volume fraction of 3.75E-02 to derive an initial evolution rate for ^{14}C -labelled gas of $6.37\text{E}+07 \text{ Bq y}^{-1}$. It has been shown that the majority of ^{14}C is not associated with cellulose; at most having an activity of 800 GBq, but more likely to be in the order of tens of GBq. This represents a direct reduction in the calculated amounts of ^{14}C -labelled gas from cellulose of between 1 and 2 orders of magnitude.

Whilst this study has focused on those waste streams contributing greater than 1% of future forecast ^{14}C activity, it is recognised that uncertainties in the waste streams contributing less than 1% of future forecast ^{14}C activity are, in some cases, significant. If actual ^{14}C activities for these waste streams were found to be close to the upper uncertainty limits stated in the 2007 National Inventory, then an additional 2 TBq of ^{14}C activity would be contributed.

VERIFICATION STATEMENT

This document has been verified and is fit for purpose. An auditable record has been made of the verification process. The scope of the verification was to confirm that:-

- The document meets the requirements as defined in the task specification/scope statement
- The constraints are valid
- The assumptions are reasonable
- The document demonstrates that the project is using the latest company approved data
- The document is internally self consistent

HISTORY SHEET

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1. Introduction

The Low Level Waste Repository (LLWR) is undertaking a programme of work leading to the production of an Environmental Safety Case by May 2011 (2011 ESC). The 2011 ESC will be submitted to the Environment Agency (EA) in order to satisfy a Requirement in the LLWR's current authorisation.

As part of work to support a submission to the Environment Agency on 1st May 2008, NNL undertook an assessment of the radiological impact of ¹⁴C-labelled gases (Ball *et al.*, 2008). This included a review of the main contributors (greater than 1%) to the ¹⁴C inventory in terms of the nature of the waste streams with which it is associated and the potential form of the ¹⁴C.

In developing the 2011 ESC, the LLWR would like to develop their knowledge of the ¹⁴C inventory in more detail and to consider a wider range of waste streams that might be associated with significant ¹⁴C. The assessment presented in this report supports an improved and more realistic consideration of the impact of ¹⁴C-labelled gas and the release of ¹⁴C via the groundwater pathway.

The objectives of this assessment were to review in detail the nature of ¹⁴C-bearing waste streams, the likely form and association of ¹⁴C within those waste streams and the potential processes that may lead to the release of ¹⁴C.

The review has been undertaken on the basis of:

- Information in the National Inventory;
- Review of Waste Stream Characterisation Documents held by the LLWR; and
- Seeking further information on key waste streams from waste consignors.

Particular focus has been given to identifying any ¹⁴C associated with cellulose-bearing wastes or present within metals as carbides or in any form that has the potential to generate ¹⁴C-labelled gases.

The review has taken account of differences induced by options for future waste disposal at the LLWR, including increased use of incineration, metal melting and/or compaction.

A review of work undertaken to date is given in Section 2, with current inventory data sources discussed in Section 3.

Section 4 presents an assessment of uncertainty in the LLWR future ¹⁴C inventory.

An examination of the potential impacts of alternative waste treatment technologies on ¹⁴C-bearing wastes and the subsequent release of activity in the near field is described in Section 5. Conclusions from this study are presented in Section 6.

It is noted that, in the process of reviewing the 2007 National Inventory source data with the Sellafield consignor, a number of errors were discovered, which revealed a significant overestimate in the forecast ¹⁴C inventory for particular waste streams. Corrected data were supplied by Sellafield Ltd for some waste streams, but it should be recognised that investigations are ongoing, and the findings of this report may need to be reviewed once the complete set of revised data is available.

2. Review of work to date

The inventory of past and potential future disposals at the LLWR (Wareing *et al.*, 2008) was calculated within the LLWR Lifetime Project both as a response to the EA's requirements under Schedule 9 of the Authorisation and as part of the preparation for the 2011 ESC. The results of the inventory calculation for ^{14}C were used as input to an assessment of the radiological impact of ^{14}C -labelled gases (Ball *et al.*, 2008). This included a review of the main contributors (greater than 1%) to the ^{14}C inventory in terms of the nature of the waste streams with which it is associated and the potential form of the ^{14}C .

The following subsections summarise the key findings from the previous work to estimate the ^{14}C inventory for use in the Requirement 2 assessment, and the subsequent ^{14}C -labelled gas pathway assessment.

2.1. LLWR ^{14}C inventory

The inventory included the following three components:

1. The Trench inventory, representing the inventory disposed to the trenches from the start of LLW operations in 1959 up to the date when trench disposals were first recorded onto the waste tracking system in July 1993.
2. The Vault 8 inventory, based on the contents of the waste tracking system database, covering from the start of Vault 8 operations until the present day. For derivation purposes, the system database was frozen for disposals at April 2007.
3. The future vaults inventory, based on the 2004 UK National Inventory and representing potential disposals to the future vaults at the LLWR.

The LLWR's waste tracking system database is used to track and locate disposals in Vault 8. With Vault 8 higher stacking in current operation, the waste tracking system database contains the full inventory of waste consigned to Vault 8 and contains information extracted directly from the disposal records, as well as additional useful information from WAMAC and LLWR operations. Information stored in the waste tracking system database has proven to be useful but is limited to describing disposals over the period 1988 to 2007. It does not therefore contain the full data-set for historic consignments to the trenches thereby requiring some conceptual modelling to complete the inventory determination.

The 2004 UK National Inventory represented the most up-to-date public domain data source at the time of calculating the inventory, providing information on radioactive waste streams with the potential to be consigned to the LLWR, both from within and without the nuclear industry.

^{14}C is an activation product present predominantly in graphite, steel and concrete arising from the decommissioning of nuclear reactors. The majority of this waste will not arise until after 2040. As such, around 95% of ^{14}C in the LLWR inventory of disposals is in the future vaults, with a total activity two orders of magnitude greater than the inventory in the trenches. ^{14}C is present in other waste types, albeit to a lesser extent, for example ion-exchange resins and organic materials such as cellulose.

The total ^{14}C inventories calculated for the trenches and vaults are as given in Table 1.

Table 1 Total ^{14}C activity contents of 2008 LLWR inventory, by disposal area

Disposal Area	Total ^{14}C activity (TBq)	Percentage of LLWR Total ^{14}C (%)
Trenches	9.98E-02	1.3
Vault 8	2.97E-01	3.8
Future Vaults	7.35E+00	94.9

The inventory of ^{14}C in the trenches was calculated from the backfitting of Vault 8 disposal records and present-day or future forecast waste streams considered representative of historical disposals. The total activity of approximately 0.1 TBq represents around 1.4% of the ^{14}C present in the future vaults. Over half of the trench ^{14}C originates from Minor Users. The Minor Users waste stream is constructed from contributions made by companies and organisations outside of the nuclear industry, including hospitals, universities and small companies that have agreements with the LLWR under the Radioactive Substances Act 1993 (RSA93) to manage and dispose of radioactive waste. These disposals are recorded as 'WS' streams, which are outwith the National Inventory publications.

The contribution to the trench ^{14}C inventory from Minor Users has been influenced by the use of a fingerprint developed from more recent data, including a few high activity level consignments that have a large influence over the averaged activity levels used to represent the historical Minor User consignments to the trenches. ^{14}C from these waste streams arises principally from two sources; reactor graphite, steel and concrete, and ion exchange materials.

The Vault 8 ^{14}C inventory was derived entirely from actual disposal data and is considered to be as accurate an assessment as can be achieved, assuming disposal records have been completed correctly and are representative of the waste being disposed. An assessment of the waste tracking system database in Ball *et al.*, (2008) identified that the majority of the ^{14}C activity in Vault 8 arose from Defence wastes and Amersham disposals. These are predominantly ion exchange resins by activity from Rosyth and Devonport and general laboratory waste from Amersham laboratories.

The future vaults ^{14}C inventory was calculated from the waste stream arisings data provided in the 2004 UK Radioactive Waste Inventory. This information consists of waste stream stock and forecast arising volumes, radionuclide fingerprint activities per unit volume and material contents. Table 2 shows those waste streams that will contribute greater than 1% of the total disposed ^{14}C activity (7.35 TBq). The principal waste stream 1A09, whilst contributing 40.7% (2.99 TBq) of the total activity, is intended for incineration, therefore it is unlikely to be disposed to the LLWR, e.g., because any residue of concentrated activity would likely be classed as intermediate level waste. Within the remaining key contributing waste streams, ^{14}C is present as an activation product in steel, graphite and concrete.

Forecast information provided for waste streams by the waste producers is subject to uncertainties, and these are discussed in more detail in Section 3.1.

Table 2 Majority contributors of ¹⁴C to the Future Vault Inventory (based on 2004 UK Radioactive Waste Inventory).

Waste Stream	Waste Stream Description	¹⁴ C waste form	Waste Volume (m ³)	Percentage of total volume (%)	¹⁴ C Activity (TBq)	Percentage of total Activity (%)
1A09	Incinerated Waste	Classed as unsuitable for LLWR disposal	3,300	1.0	2.99E+00	40.7
5F303	Windscale Pile 2 LLW	Present as activation product in steel and concrete	2,430	0.7	2.09E+00	28.4
5F301	Windscale Pile 1 LLW	Insoluble ¹⁴ C from activated steel and concrete	1,768	0.5	4.60E-01	6.3
7D39/C	LLW Submarine Ion Exchange Resin	Vast majority of the ¹⁴ C has been removed by the pre-conditioning that has been undertaken. IX Resin decayed from ILW.	99*	0.0	4.04E-01	5.5
5G303	Dragon Reactor Decommissioning LLW	Carbon and stainless steel (68%), graphite (2%), reinforced concrete/rubble (29%) and plastics (1%).	1,792	0.5	3.58E-01	4.9
7E26	Low Level Resin from Submarines & Effluent Treatment Plant	¹⁴ C is present in metal salts e.g., carbonates	27	0.0	1.41E-01	1.9
1A02	LLW Non-Compactable Drummable	Laboratory refuse	4,650	1.4	1.34E-01	1.8
1A03	LLW Non-Compactable Non-Drummable	Laboratory refuse	4,650	1.4	1.34E-01	1.8
7V26	Vulcan Supercompactable Drummed Low Level Waste	Present but chemical form not assessed. A varied soft trash waste stream	155	0.0	1.08E-01	1.5
5F307	Windscale Advanced Gas-Cooled Reactor (WAGR) Decommissioning LLW	¹⁴ C is present as activation product within steel and concrete.	5,480	1.6	1.05E-01	1.4
3S06	Spent Resins (LLW)	Trace quantities may exist adsorbed onto the resin beads. Detailed analysis still awaited	421	0.1	9.27E-02	1.3
1B09	Very Low Level Waste	¹⁴ C labelled organic compounds.	4,950	1.5	7.72E-02	1.1

*Packaged Waste Volume

Whilst the calculated annual future ¹⁴C inventory up to 2055 was shown to be consistently above the current annual LLWR site activity limit for ¹⁴C up to 2055, it was also shown that LLW streams of significantly higher ¹⁴C activity (over an order of magnitude higher in some cases) are forecast to arise beyond 2075. These higher-activity ¹⁴C-bearing waste streams are predominantly activated reactor graphite moderators, together with activated reactor steel and concrete. Future strategy for the management of UK LLW is seeking to increase the longevity of the LLWR such that it may be in operation post-2075; therefore high ¹⁴C-bearing streams arising during this period should be taken into consideration.

2.2. Radiological assessment of ¹⁴C-labelled gas

Ball *et al.*, (2008) included a review of previous information, which resulted in an update to calculations assessing impacts arising from the gaseous transport of ¹⁴C to the biosphere and its subsequent incorporation and ingestion in food crops. Table 3 shows the subsequent ¹⁴C concentration in plants due to release of ¹⁴C-labelled gases from the LLWR and the corresponding calculated effective dose.

The review included an update to the ¹⁴C inventory based on the most recent calculations (Wareing *et al.*, 2008), a review of the processes of ¹⁴C release, consideration of DRINK model results and a review of the RIMERS-based dose assessment model developed for the UK Nirex Ltd assessment (Nirex, 1994; 1997).

Table 3 Annual Effective Doses from ¹⁴C-labelled gas (Ball *et al.*, 2008)

Location	¹⁴ C Plant Concentrations (Bq kg[C] ⁻¹)	Annual Effective Dose (Sv)
Trenches	19	4.1E-07
Vault 8	142	3.0E-06
Future Vaults	205	4.4E-06

The 2002 assessment of ¹⁴C release to the groundwater pathway (using the DRINK model) and the gas pathway (McGarry, 2003) made the conservative assumption that all ¹⁴C was associated with cellulosic materials. Further evaluation of the inventory, particularly of the future vaults, indicated that cellulosic materials actually comprise a minor proportion of the wastes. Waste stream 1A09 "incinerated waste" comprises some biodegradable cellulose materials (14 wt%) with the potential to produce CH₄ gas that could migrate from the cementitious vault. Other materials in this waste stream include organic liquids and plastics (35 wt%). These organic substances would be expected to have a lower rate of biodegradation than cellulose (BNFL, 2002) and thus would have a lower release of ¹⁴C in gaseous form. Disposal of the organic liquids in this waste stream would require treatment to be suitable for disposal e.g. encapsulation in cement. If the material were incinerated, the active residue is unlikely to be suitable for LLW disposal, as noted in the 2004 UK National Inventory. As noted above, it is unlikely that 1A09 will be disposed to the LLWR following incineration, due to an increase in activity concentration to levels beyond those acceptable under the current LLWR Conditions for Acceptance (CFA).

Other major ¹⁴C active waste streams listed include:

- Activation products in steel and concrete from decommissioned reactors (e.g., 5F303, 5F301, 5F307) comprise around 36% of the inventory. In the case of steel, ¹⁴C release will be dependent on the corrosion of the steel. The chemical form of the ¹⁴C in activation products in metals is uncertain (Small, 2004) and could be either organic (potentially yielding CH₄) or inorganic, in which case it is likely to be immobilised in carbonate corrosion products and carbonation of the backfill grout. Activation product ¹⁴C within concrete (excluding steel rebar) is more likely to be inorganic.
- Ion exchange resins (e.g., 7D39/C, 7E26, 3S06) comprise around 10% of the ¹⁴C future vaults inventory. In these wastes it is noted in the description that ¹⁴C is present as carbonate salts. The LLWR CFA require that these wastes are encapsulated in cement grout, which would reduce the solubility and mobility of ¹⁴C.
- Miscellaneous laboratory refuse (1A02, 1A03 which are noted to be non compactable and thus unlikely to comprise paper materials), and VLLW organic

compounds (1B09) which together comprise less than 5% of the future vault inventory.

It is unlikely that ^{14}C in the waste streams contributing the greatest activity (e.g. where ^{14}C is present as an activation product or in ion exchange resins) would give rise to ^{14}C -labelled methane. The assumption in the 2002 PCSC that all ^{14}C is associated with cellulosic material is very pessimistic. A more realistic assessment could be undertaken based on the observation that only less than 1% of ^{14}C is associated with cellulosic material.

Table 4 shows the calculation undertaken in Ball *et al.*, (2008) to derive the effective dose and hence risk arising from the release of ^{14}C -labelled gas from the LLWR future vaults. The bases for these calculations and parameters, taken from Ball *et al.*, (2008), are provided below.

To determine evolution rates for ^{14}C -labelled gases, the amount of ^{14}C associated with cellulosic waste materials in relation to all constituents of the waste was considered. As the exact amount of ^{14}C associated with cellulose is unknown, it was assumed that all gaseous ^{14}C evolution was from the cellulosic volume fraction of the waste. This approach effectively uses the volume fractions to scale the component future vault ^{14}C inventory of 7.35 TBq. Grout and container metal associated with the vault waste were considered not to be contaminated and were excluded from the calculations. The cellulosic volume fraction was calculated as 0.0375 for the future vaults.

Ball *et al.*, (2008) states that a reasonable estimate for a half life $T_{0.5 \text{ gas}}$ (y) for the DRINK model output was considered to be 250 y. Allowing for a contribution from the decay of ^{14}C , a total degradable fraction, DF, of $2.89\text{E-}3 \text{ y}^{-1}$ was derived using Equation 1, comprising $2.77\text{E-}3 \text{ y}^{-1}$ for gas production and $1.21\text{E-}4 \text{ y}^{-1}$ for ^{14}C decay ($T_{0.5 \text{ decay}} = 5730 \text{ y}$).

$$\text{DF} = \log_e 2 / T_{0.5 \text{ gas}} + \log_e 2 / T_{0.5 \text{ decay}} \quad \text{Equation 1}$$

Initial Gas Evolution Rates, IER, (Bq y^{-1}) for each facility component were derived using Equation 2.

$$\text{IER} = I * \text{vf} * \text{dfc} * \text{grf} * \text{DF} \quad \text{Equation 2}$$

where:

I is the component inventory (Bq)

Vf is the component cellulosic volume fraction (dimensionless)

dfc is the degradable fraction of cellulose (dimensionless): = 0.8

grf is the gas release fraction (dimensionless): = 0.1 (vaults)

DF is the degradable fraction (y^{-1}).

The specific activity of ^{14}C in plants was derived using the initial release rate per unit area with an estimate of the mass of ^{14}C in plants from a unit release per unit area. From the updated and re-parameterised RIMERS model, a chronic release of $1 \text{ Bq m}^{-2} \text{ d}^{-1}$ to the microbial biomass compartment resulted in an equilibrium ^{14}C concentration in standing biomass of 67.9 Bq m^{-3} of carbon dioxide at Standard Temperature and Pressure (STP). As 1 m^3 of carbon dioxide contains 44.64 moles of gas, a mass of 0.536 kg[C] can be derived. In terms of specific activity, this equates to $127 \text{ Bq kg[C]}^{-1}$ for the standing biomass.

On the basis of the specific activity being scaled to the unit chronic release expressed over a year and multiplying by the initial release rate per unit areas, the ^{14}C plant concentrations Bq kg[C]^{-1} were derived, as shown in Table 4.

The plant concentrations are based directly on a correlation between ^{14}C activity in the waste and the amount of ^{14}C -labelled gas generated. Section 4 discusses, in more detail than Ball *et al.*, (2008), the quantities of cellulosic materials expected to be consigned to the future vaults, and how much of the cellulosic material is likely to contain ^{14}C activity.

Table 4 Impact of updated ^{14}C inventory on potential dose uptake from future vaults disposals

Gas evolution rates					
Degradable Fraction Of Cellulose	Gas Release Fraction	Degradation Rate Of This Degradable Fraction	Radioactive Decay Constant	DF	Specific Activity
(-)	(-)	(y^{-1})	(y^{-1})	(y^{-1})	($\text{Bq kg}[\text{C}]^{-1}$)
0.8	0.1	2.77E-03	1.21E-04	2.89E-03	127

Release Rates					
Future Vault Inventory	Future Vault Area	Cellulosic Volume Fraction	Initial Evolution Rate	IER per unit area	Plant Concentration
(Bq)	(m^2)	(-)	(Bq y^{-1})	($\text{Bq m}^{-2}\text{y}^{-1}$)	($\text{Bq kg}[\text{C}]^{-1}$)
7.35E+12	107,849	3.75E-02	6.37E+07	591	205.66

Dose Rates					
	Plant Concentration	Annual Intake	Effective Dose Intake	Effective Dose	Risk
	($\text{Bq kg}[\text{C}]^{-1}$)	($\text{kg}[\text{C}]\text{y}^{-1}$)	(Sv Bq^{-1})	(Sv y^{-1})	(y^{-1})
Adult	206	36.525	5.80E-10	4.4E-06	3E-07
Child	206	24.35	8.00E-10	4.0E-06	2E-07
Infant	206	12.175	1.60E-09	4.0E-06	2E-07

3. Current ¹⁴C inventory data sources

The ¹⁴C inventory used as a basis for the Requirement 2 calculations was, as discussed above, based on the 2004 UK Radioactive Waste Inventory, which was the most up-to-date publicly-available information at the time. However, the 2007 UK Radioactive Waste Inventory (Pöyry, 2008) is now available. This more-recent information post-dates the Requirement 2 inventory data by up to three years and therefore may influence and change any prioritised list of waste streams for further investigation. Previous comparative studies of National Inventories have shown some significant differences in waste stream data between successive publications.

For waste streams currently disposed to the LLWR, additional detailed inventory data may be available within the LLWR Waste Stream Characterisation Documents (WSCDs); documentation required to be provided by the waste producers to LLWR prior to agreement that waste will be accepted for disposal.

In this study the following information sources have been used:

- 2007 UK Radioactive Waste Inventory;
- Waste Stream Characterisation Documents; and
- Other information sought directly from the waste producers.

The following subsections discuss these data sources with regards to information on ¹⁴C-bearing wastes in more detail.

3.1. 2007 UK Radioactive Waste Inventory

The 2007 UK Radioactive Waste Inventory is the latest in a series of publicly-available National Inventory reports commissioned by the UK Government, providing comprehensive and transparent information on UK radioactive wastes.

The National Inventory is a snapshot of radioactive waste in the UK as at 1st April 2007. It is based on a single scenario for the production of radioactive waste in the UK. This scenario reflects the status of UK nuclear industry and Government policies at that date. For example, the inventory assumes that no new nuclear power stations will be built in the future in the UK and that radioactive materials such as spent nuclear fuels are not classified as waste, as they could be reused in the future.

Raw arising and packaged volumes are given for the UK as a whole, for each type of waste-producing activity and for each of the main waste-producing organisations. The report also presents information on the radioactivity and the material content of the wastes.

The report includes high-, intermediate- and low-level wastes produced from uranium enrichment, nuclear fuel manufacture, nuclear power production, spent fuel reprocessing, research and development, medical and industrial sources and defence activities.

The radionuclide fingerprint tables provided in the 2007 National Inventory give specific activities in terabequerels per cubic metre. Codes are associated with each radionuclide activity to show the upper and lower uncertainty bands and to indicate how activities have been derived.

Future waste stream radionuclide fingerprints and volumetric estimates submitted to the UK National Inventory are derived using a variety of methods, dependent on the information available to the waste producer at the time of submission. Activities and volumes for current wastes may be directly measured whereas, for wastes arising in the

future, estimates may be provided. This has a direct influence on the confidence in radionuclide activity data, with measured values liable to have greater accuracy than estimated values.

Similar to the activity data, the volumetric data provided in the National Inventory are subject to differing derivation methods, dependent on whether waste is currently stored or arising, or is forecast to arise at a future date. Typically, volumes may be derived through direct measurement, calculated from process flow-sheets, or for decommissioning wastes, estimated from building dimensions and/or observation.

Comments are provided occasionally by the waste producers in reference to confidence in future forecast volumes and the method of derivation, but uncertainty bands are not assigned to the volumetric data.

Material compositions are given in the National Inventory as percentage weights or volumes for each waste stream. Data quality is highly variable. Addition of the given material contents for a waste stream may result in totals equal to, less than or greater than 100%. Totals greater than 100% are obtained where there is uncertainty in the material composition and a number of materials are assigned values with a 'less than' prefix.

As has been observed in the past with successive National Inventories, a high level comparison of the 2004 and 2007 UK Radioactive Waste Inventories shows a number of differences. Specifically for ^{14}C -bearing wastes:

- The total inventory of ^{14}C in LLW the 2004 UK National Inventory was 111 TBq, compared with 43 TBq in the 2007 UK National Inventory (of which 1.7 TBq is from all waste streams designated for consignment to the LLWR);
- The waste streams 9C316 and 9F317 (Final Dismantling & Site Clearance Graphite LLW from Dungeness A and Sizewell A respectively) have been removed; and
- 1A02 and 1A03 (Non-compactable LLW from GE Healthcare) contained 268 GBq of ^{14}C in the 2004 UK National Inventory and now in the 2007 UK National Inventory contain zero ^{14}C activity.

The implications of these changes on the calculated LLWR ^{14}C inventory are discussed in more detail in Section 4.

3.2. Waste Stream Characterisation Documents

WSCDs are required to be produced for every waste stream routed for LLWR disposal. The documents are valid for a three-year period, after which they must be reviewed and reissued. WSCDs must provide, as a minimum, the following information:

- Unique waste stream Identifier;
- Origin of the waste including the processes that created the waste;
- Physical composition of the waste;
- Radioactive composition of the waste;
- Processing of waste;
- Packaging of the waste; and
- Volume to be treated / disposed.

The WSCDs include a description of the methodologies used in deriving the inventory data, and therefore are likely to be of use in providing additional information not given in the 2007 UK Radioactive Waste Inventory.

3.3. Further information

A detailed assessment of the National Inventory forecasts was required for a number of key waste streams to underpin the ^{14}C inventory estimate, explain how the ^{14}C may be distributed through the waste materials and to understand potential treatments the waste may undergo prior to final packaging for disposal. This additional information came from three main sources:

- Waste Stream Characterisation Documents;
- Discussions with the waste producers; and
- Evaluation of Waste Receipt Monitoring (WRM) undertaken at WAMAC, Sellafield.

For waste streams of significance where WSCDs are not available – i.e. those waste streams not forecast to arise within the next three years - calculations or assumptions underpinning the published National Inventory may be obtained to assist in understanding the uncertainties surrounding the LLWR ^{14}C inventory. This additional information was obtained directly from the waste producers. This approach was also taken where an assessment of WSCDs for existing waste streams did not yield information of sufficient clarity.

Waste Receipt Monitoring records are available from the Waste Monitoring and Compaction Plant (WAMAC) at Sellafield. These show actual activities measured using a High Resolution Gamma Scanner (HRGS) for selected waste streams received for processing at WAMAC. These data were examined to identify potential differences between measured and declared activities for ^{14}C -bearing wastes.

Table 5 shows how additional information was sought from each waste producer for prioritised waste streams.

Table 5 Action undertaken by NNL to source further information

Site	Action
Sellafield	Reviewed prioritised WSCDs and held discussions with the waste stream compilers.
Devonport	Reviewed prioritised WSCDs and held discussions with the waste stream compilers.
Windscale	Discussions were held with waste stream compilers.
Magnox South	Discussions were held with waste stream compilers.
Magnox North	Discussions were held with waste stream compilers.
Winfrith	WSCDs were reviewed prior to discussions with the appropriate waste stream data compilers.
GE Healthcare Ltd	Reviewed WSCDs.

4. Assessment of LLWR ¹⁴C inventory uncertainty

It was determined that the assessment of uncertainty in the LLWR ¹⁴C inventory of disposals should be focused primarily on the future vaults, as most of the ¹⁴C inventory is associated with future disposals. In addition, the assessment would take particular account of the physical makeup of ¹⁴C-bearing wastes in order to provide an understanding of the distribution of ¹⁴C activity through different material types and hence inform a study of release rates through groundwater and gaseous pathways.

The assessment began with a review of the 2007 National Inventory dataset to identify ¹⁴C-bearing waste streams. These waste streams were compared with the waste streams contributing ¹⁴C in the 2004 National Inventory to identify differences from the dataset used as a basis for in the Requirement 2 submission. An appraisal was undertaken to ascertain how activity is distributed across different material types. From this, key waste streams considered to be of most influence were identified; in particular those waste streams with materials and/or waste-form considered of most interest.

Further information was sought from the WSCDs, waste producers and the WRM records in order to cross-check National Inventory data and improve understanding of specific issues relating to waste characterisation.

The following sub-sections discuss these steps in more detail.

4.1. Review of 2007 National Inventory data

The 2007 National Inventory data were acquired in Microsoft Access database form to enable manipulation. Radionuclide fingerprints were assessed to identify those waste streams containing ¹⁴C. Annual arising activities in GBq were calculated for each waste stream and summed for each year to give a total annual ¹⁴C arising activity for all LLW in the 2007 National Inventory (Figure 1).

A total of 42,500 GBq of ¹⁴C is forecast to arise in UK LLW up to 2129. It can be seen in Figure 1 that the large majority of ¹⁴C in LLW (~95%) is forecast to arise beyond 2076. These arisings are predominantly decommissioning wastes from reactor sites, mainly comprising activated structural steel, concrete and moderator graphite. ¹⁴C-bearing waste streams with current stocks or arisings in the immediate future have been identified; these being a mixture of operational and decommissioning waste streams from a range of sites. Whilst these waste streams are generally of lower activity than later arisings, they may nevertheless be of significance in terms of their potential impact on the LLWR.

Figure 2 shows the total annual ¹⁴C arising activity for all LLW split by the stated waste stream disposition routes as given in the 2007 National Inventory. It can be seen that the wastes arising beyond 2076, which account for the majority of ¹⁴C arisings, are designated as either 'Not expected to be disposed of to LLWR' or 'Unsuitable for disposal to LLWR' (terms as stated in the 2007 National Inventory).

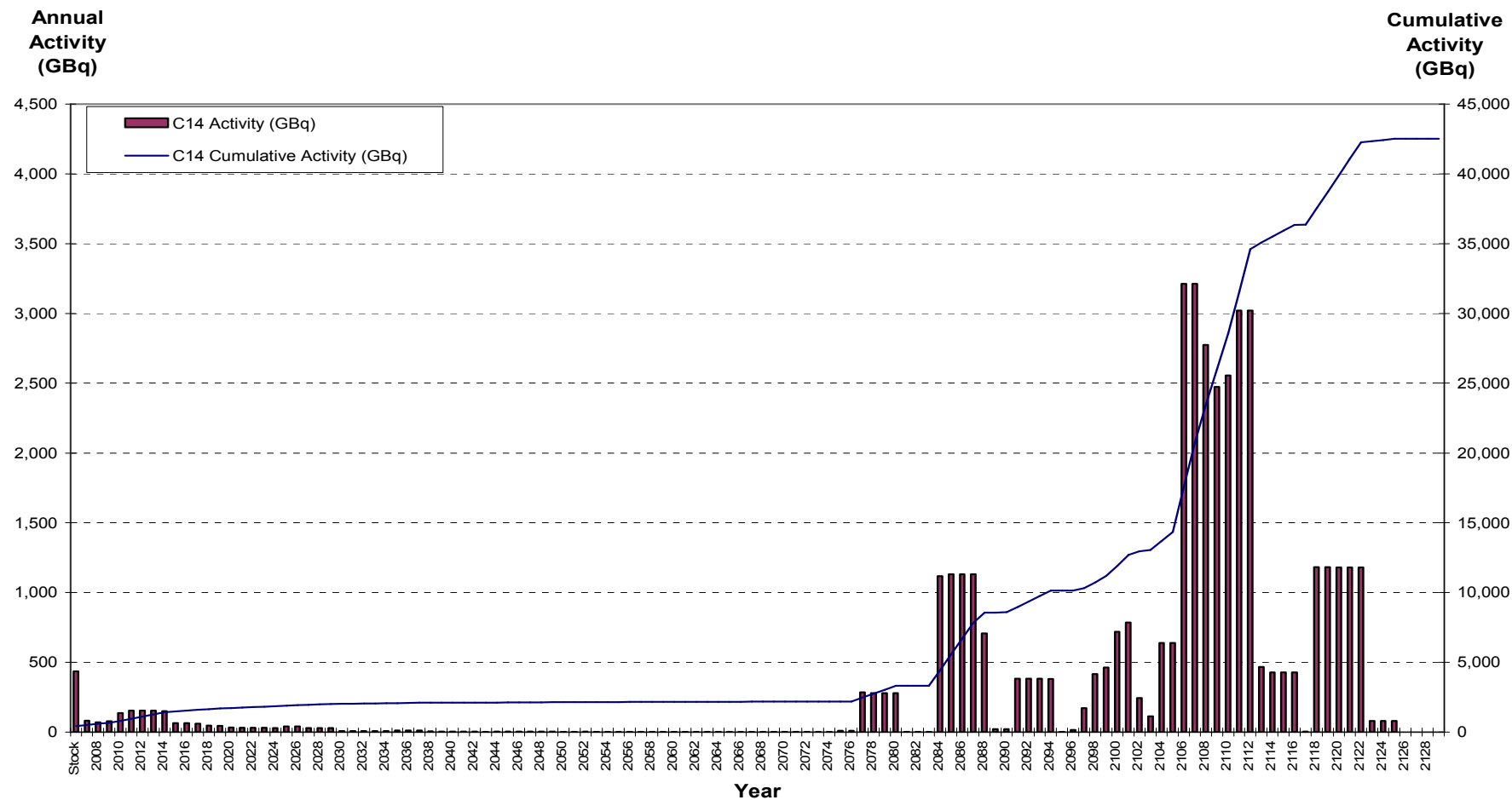


Figure 1 ¹⁴C present in LLW in the 2007 National Inventory

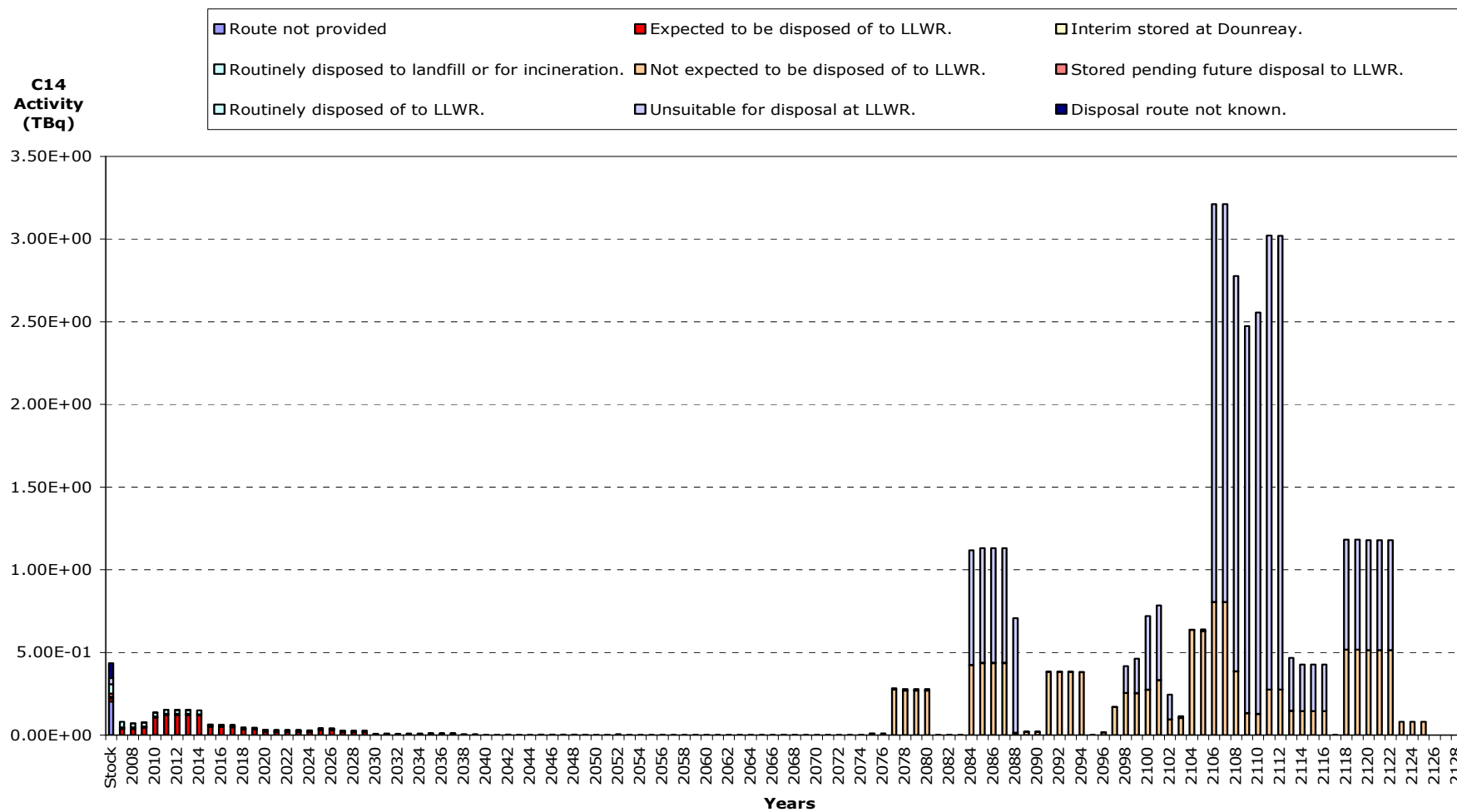


Figure 2 ¹⁴C by each LLW disposition route in the 2007 National Inventory

It should be noted, however, that the waste streams designated 'Not expected to be disposed of to the LLWR' are not necessarily unacceptable for LLWR disposal in terms of their meeting the current Conditions for Acceptance (CFA). Guidance given to waste producers in completing questionnaire returns for input to the 2007 National Inventory (Nirex Report N/135) states "*The LLWR has a finite volumetric and radiological capacity; it should be assumed that disposals would stop in 2050. For LLW streams that arise after 2050 you should respond: 'Not expected to be consigned to the LLWR' "*. These waste streams could be of potential significance to the LLWR should operations be extended beyond 2050.

The significant majority of ^{14}C -bearing waste streams arising in the period up to around 2040 are designated as either 'Expected to be disposed of to the LLWR' or 'Routinely disposed of to the LLWR'. Whilst these waste streams represent only around 5% of forecast future ^{14}C activity arisings, it is important to note that annual forecasts in this period are close to, or above, current LLWR annual limits for ^{14}C .

Table 6 shows the top fifty ^{14}C -bearing waste streams in the 2007 National Inventory that are forecast for disposal at LLWR, ranked in descending order of total ^{14}C activity. The table shows that the total ^{14}C inventory is now 1.7 TBq; a 4-fold reduction from the inventory used as the basis for the Requirement 2 submission.

It is noted that the activities shown are as given in the 2007 National Inventory. Subsequent discussion with Sellafield Ltd has revealed a number of significant overestimates in the data for Sellafield '2X' waste streams, which are currently under investigation. The assessment described in Section 4 uses partially corrected data; i.e. those 2X waste streams in the top half of Table 6 have been revised based on new information from Sellafield Ltd. The remaining Sellafield streams are not significant in terms of their ^{14}C content but will be reported following work currently being undertaken by Sellafield Ltd.

A number of waste streams are highlighted in Table 6. The first seven waste streams (7D39/C to 2C920) are forecast to contribute around 75% of future ^{14}C arisings in waste streams designated for disposal at LLWR. Of these, the top two streams 7D39/C, Devonport Submarine Ion Exchange Resin, and 9E958, Oldbury Care & Maintenance Preparation Dry Fuel Route, contribute around 25% and 20% respectively; thus accounting for almost half of all future forecast ^{14}C arisings for LLWR waste streams.

Two further waste streams lower down the rankings are highlighted: 1B02, GE Healthcare Cardiff Non-compactable Drummable; and 7E23, Rosyth Royal Dockyard Metallic Waste. Both these streams have contained significant ^{14}C activity in their disposals to LLWR Vault 8 in the past (approximately 34 and 20 GBq respectively) but GE Healthcare Ltd forecast these streams to contain only a fraction of this ^{14}C activity in the future arisings.

Waste streams towards the bottom of the rankings in Table 6 each contribute only around 0.1% of forecast future ^{14}C arisings for LLWR disposal.

The waste stream volumes and activity uncertainty bandings shown in Table 6 are of interest. Where waste streams have a high forecast arising volume and a high uncertainty band then application of the upper uncertainty band could have a significant effect on the total activity contribution. For example, 2C920, Chapelcross Care & Maintenance Prep (Reactor LLW) has an arising volume of 43,132 m³ and an upper uncertainty band of C (a factor of 10). If the upper uncertainty band were to be applied then the total ^{14}C activity contribution from this waste stream would rise from 86 GBq, representing around 4.9% of total future arisings, to 860 GBq, which would represent around 34% of a significantly larger total. It is not currently understood how the compilers estimate or calculate the confidence bands for the radionuclide activity in their waste streams.

Table 6 Waste streams forecast to contain the majority of ¹⁴C in the LLWR future vaults

Order	¹⁴ C activity (GBq)	Waste stream	% of Total ¹⁴ C in LLW	Raw volume (m ³)	Packaged volume (m ³)	First year of arisings	Final year of arisings	Upper banding (stock / arising)	% increase in total ¹⁴ C inventory when upper banding applied	Activity received by the LLWR pre 2007 (GBq)
1	442	7D39/C	25.0%	108	293	Stock	2029	B/B	50%	
2	355	9E958	20.1%	5,076	6,041	2008	2019	/C	181%	
3	119	5G303	6.7%	2,143	2,996	2007	2014		Unknown	
4	111	3S06	6.3%	506	2,738	Stock	2037	C/C	57%	9.06
5	105	5F307	5.9%	5,480	9,716	2007	2014	/B	12%	2.75
6	104	2X927	5.9%	4,199	5,049	2007	2113	/C	53%	0.00
7	86	2C920	4.9%	43,132	70,089	Stock	2017	C/C	44%	
8	33	7E26	1.9%	6	42	Stock	2028		Unknown	19.88
9	28	2X21	1.6%	2,050	1,859	2007	2025	/C	14%	0.04
10	24	2C13	1.4%	1,224	2,170	Stock	2007	A/A	1%	0.18
11	22	9C911	1.2%	2,438	2,417	2009	2020	/C	11%	
12	20	2D109	1.1%	67,302	55,515	2012	2095	/C	10%	
13	20	2X31	1.1%	1,394	924	2007	2041	/C	10%	0.22
14	19	7D26/C	1.1%	166	718	Stock	2029	A/A	1%	
15	18	2C921	1.0%	9,094	14,777	Stock	2017	C/C	9%	
16	17	6H02	1.0%	8,600	11,781	2007	2049	/C	9%	
17	13	3M110	0.7%	431	469	2024	2031	/C	7%	
18	13	4C110	0.7%	423	461	2025	2032	/C	6%	
19	13	1B03	0.7%	660	825	2007	2039	/A	0%	0.85
20	12	9E914	0.7%	1,160	1,380	2010	2017	/C	6%	
21	11	5G301	0.6%	6,666	8,657	2007	2013	/B	1%	0.00
22	10	9F910	0.6%	4,870	4,934	2009	2018	/C	5%	
23	9	9E960	0.5%	88	75	2010	2014	/B	1%	
24	8	2X140	0.5%	670	771	2012	2089	/C	4%	0.00
25	8	3S301	0.5%	8,131	4,719	2044	2053	/C	4%	
26	8	7E28	0.5%	2	14	Stock			Unknown	9.32
27	6	2C922	0.4%	3,243	5,269	2010	2017	/C	3%	
28	6	2X49	0.3%	13,603	5,493	2007	2119	/C	3%	0.00
29	6	5H304	0.3%	1,793	2,281	2014	2016	/A	0%	
30	5	5C51	0.3%	16	30	Stock		/A	0%	
31	5	9E104	0.3%	172	112	2023	2108	/C	3%	
32	5	7E101	0.3%	173	221	Stock	2028	A/A	0%	
33	4	2C924	0.3%	2,235	3,631	Stock	2017	A/A	0%	
34	4	9E58	0.2%	61	65	Stock	2014	B/B	0%	1.50
35	4	7D33	0.2%	5	2	2007	2029	/C	2%	
36	4	9A105	0.2%	124	81	2012	2073	/C	2%	
37	4	1B02	0.2%	33	21	2007	2039	/C	2%	34.11
38	4	7F26/C	0.2%	74	199	2007	2099	/A	0%	
39	4	9G105	0.2%	354	575	Stock	2011	B/B	0%	7.71
40	4	3L11	0.2%	173	174	Stock	2016	C/C	2%	1.02
41	3	9C105	0.2%	170	111	2021	2101	/C	2%	
42	3	3S303	0.2%	735	919	2045	2053	/C	2%	
43	3	3S101	0.2%	97	82	2041	2056	/C	1%	
44	3	9C13	0.2%	72	68	Stock	2009	C/C	1%	0.10
45	3	2C11	0.2%	1,437	1,186	Stock	2009	A/A	0%	0.73
46	2	9C912	0.1%	830	1,090	2009	2020	/C	1%	
47	2	9H922	0.1%	825	1,341	2012	2020	/C	1%	
48	2	7E23	0.1%	238	324	Stock	2028	C/C	1%	20.08
49	2	9J949	0.1%	3,760	4,587	Stock	2016	C/C	1%	0.32
50	2	9C11	0.1%	112	120	Stock	2009	C/C	1%	0.58
Total	1,764				723,699	Stock	2119			278.1

Note: Factor change provided by waste stream compiler for upper confidence values. Two letters are provided where both stocks and arisings are present.

A	B	C	D
1.5	3	10	100

4.2. Identification of key waste streams

In order to focus effort on the most significant waste streams, a subset of waste streams was identified for further study. Further information was obtained on each of these waste streams from the National Inventory, WSCDs and the waste producers. Waste streams were identified on the basis of total future lifetime ^{14}C activity content, waste form, potential for release of ^{14}C to groundwater or as ^{14}C -labelled gas and uncertainty in activity concentration. In addition, waste streams exhibiting significant ^{14}C content in past consignments but forecasting insignificant future ^{14}C content were identified.

Table 7 lists the 23 waste streams identified for further investigation. Of these, 17 are in the top 20 waste streams contributing ^{14}C to LLWR future disposals (Table 6), accounting for approximately 87% of future forecast ^{14}C activity; 5 are waste streams that have consigned significant amounts of ^{14}C to Vault 8 but are forecast to contribute insignificant or no ^{14}C in the future. 2N05/C has been included as a stock within the National Inventory to represent wastes already consigned to Vault 8.

A qualitative evaluation of the data has been undertaken in order to understand the potential impact of ^{14}C in each waste stream, both in terms of the release of ^{14}C -labelled gases and on the groundwater pathway. This assessment was carried out prior to discussions with the waste producers. Any further information that may have bearing is recorded in the later tables in Section 4.5.

Table 7 ¹⁴C-bearing waste streams identified for further investigation

Waste Stream	% of ¹⁴ C in LLWR	Waste Stream Description	Waste Form (as given in 2007 National Inventory)	¹⁴ C release potential to gas	¹⁴ C release potential to GW	NNL Comment on materials present within the waste stream	NNL Comment on Inventory and Upper Bound Uncertainty of the waste stream
7D39/C	25.0	LLW Submarine Ion Exchange Resin	The vast majority of the ¹⁴ C has been removed by the pre-conditioning that has been undertaken.	High	Low	7D39/C is a conditioned waste stream comprising ion exchange resin encapsulated in cement. This process will not remove ¹⁴ C; however it should ensure reaction of ¹⁴ CO ₂ released, thus groundwater release potential is low. Potential exists for the resin to degrade and liberate ¹⁴ CH ₄ . ¹⁴ C present on the resin is likely to be in the form of carbonate, thus the nature of the resin degradation process requires consideration.	7D39/C is a conditioned stream containing encapsulated ion exchange resin. It represents about 25% of the ¹⁴ C in the future vaults and has an upper band B, which if applied could increase the ¹⁴ C in the future vaults by 50%. The presence of concrete in the waste gives a high possibility of the waste form generating ¹⁴ CO ₂ and ¹⁴ CH ₄ . Description of the characterisation and volume estimate methodologies required. The stream results from treatment of the LLW Stream 7D41.
9E958	20.1	Care & Maintenance Preparation : Dry Fuel Route (excluding BCD) LLW	¹⁴ C may be present as contamination in the form of graphite dust.	High	Medium	Graphite dust is associated with a small amount of cellulosic material. As it is present as a dust then the graphite may be subject to microbial attack. Both ¹⁴ CH ₄ and ¹⁴ CO ₂ could potentially be released from this waste. Given that no cementitious material is intimately associated with the waste then the potential for carbonation may be reduced. Further information on the cellulose present and the presence of cement would be useful.	9E958 from Oldbury is expected to be disposed of to the LLWR and contains about 20% of the future vaults ¹⁴ C activity. There is a large upper bound value associated with the waste stream, which if applied, could increase the future vaults inventory by just under a factor of 2. The ¹⁴ C is present in the waste form as graphite dust. Assessment of the WSCD to possibly determine a more focused confidence band in the forecast. If the graphite dust is fully present amongst the cellulose (3% of the material composition), is it an issue?
5G303	6.7	Dragon Reactor Decommissioning LLW	Radionuclides are present as trace elements in the activated materials.	Medium	Medium	¹⁴ C is present as an activation product in mild steel. Release through corrosion may liberate ¹⁴ CH ₄ or ¹⁴ CO ₂ . Potential exists to generate H ₂ gas, but this may be consumed by biogeochemical processes. Given low cement content ¹⁴ CO ₂ and carbonate may permit release of ¹⁴ C to groundwater.	5G303 is expected to be disposed to the LLWR and contains Dragon reactor decommissioning wastes from Winfrith. No upper bounding was supplied by the waste compiler for the ¹⁴ C component of the waste stream.
3S06	6.3	Spent Resins (LLW)	Trace quantities may exist adsorbed onto the resin beads. Detailed analysis is still awaited.	High	High	This waste stream does not contain cement and thus potential that H ¹⁴ CO ₃ ⁻ could be released to groundwater. Potential exists for ¹⁴ CH ₄ and ¹⁴ CO ₂ gas generation through degradation of the resin. Should confirm the presence of any cement encapsulation as a treatment prior to disposition.	Assessment of previous 3S06 consignments to Vault 8 have shown that the ¹⁴ C activity is approximately an order of magnitude lower than that declared in the 2007 UK National Inventory. Encapsulation is listed as the treatment but is not included in the materials.

Waste Stream	% of ¹⁴ C in LLWR	Waste Stream Description	Waste Form (as given in 2007 National Inventory)	¹⁴ C release potential to gas	¹⁴ C release potential to GW	NNL Comment on materials present within the waste stream	NNL Comment on Inventory and Upper Bound Uncertainty of the waste stream
5F307	5.9	Windscale Advanced Gas-Cooled Reactor (WAGR) Decommissioning LLW	¹⁴ C is present as activation product within steel and concrete.	Medium	Low	Potential exists for ¹⁴ CH ₄ and ¹⁴ CO ₂ release during the corrosion of activated metals, however metal is a minor component. ¹⁴ CO ₂ present in irradiated concrete will be immobile in the form of CaCO ₃ . Given the cellulose content, there is the potential for methane release and further carbonation of the concrete.	¹⁴ C concentration as stated in the disposal records is approximately an order of magnitude lower than declared in the 2007 National Inventory.
2X927	5.9	Wheelabrator Operations LLW	Not stated	Medium	Low	Potential exists for ¹⁴ CH ₄ and ¹⁴ CO ₂ release during the corrosion of activated metals, which are a significant component thus higher potential for ¹⁴ CH ₄ gas release than 5F307. ¹⁴ CO ₂ present in irradiated concrete will be immobile in the form of CaCO ₃ . Given the cellulose content potential exists for methane generation and further carbonation of the concrete.	Activity concentration appears to be orders of magnitude higher for ¹⁴ C than has already been disposed to Vault 8 for this stream. (10 ⁻⁹ vs 10 ⁻⁵ TBq/m ³). Note that this waste stream has been identified by Sellafield Ltd as having an incorrect fingerprint in the National Inventory (Khan <i>et al.</i> , 2009).
2C920	4.9	Care and Maintenance Preparation (Reactor LLW)	The chemical form of ¹⁴ C has not been determined.	Medium	Low	Potential exists for ¹⁴ CH ₄ and ¹⁴ CO ₂ release during the corrosion of activated metals, which are a significant component thus higher potential for ¹⁴ CH ₄ gas release than 5F307. ¹⁴ CO ₂ present in irradiated concrete will be immobile in the form of CaCO ₃ . Given the cellulose content potential exists for methane generation and further carbonation of the concrete.	2C920 has been given the upper band C, which denotes an order of magnitude uncertainty in its estimate of ¹⁴ C activity present, potentially increasing the inventory expected to be disposed to the LLWR by about 44%.
7E26	1.9	Low Level Resin from Submarines & Effluent Treatment Plant	¹⁴ C is present in metal salts e.g. carbonates.	High	High	This waste stream does not contain cement and thus there is potential that H ¹⁴ CO ₃ ⁻ could be released to groundwater. Potential exists for ¹⁴ CH ₄ and ¹⁴ CO ₂ gas generation through degradation of the resin. The presence of any cement encapsulation (as 3S06) should be confirmed.	No uncertainty banding has been declared in the UK National inventory for this stream; however comparable quantities of ¹⁴ C have been consigned to the LLWR in Vault 8.
2X21	1.6	Magnox Ponds West: Magnox Flask Maintenance LLW	Not stated	Medium	Medium	Potential exists for ¹⁴ CH ₄ and ¹⁴ CO ₂ release during the corrosion of activated metals. There is the potential for gas to be generated from cellulose and other organics. The release of ¹⁴ C to groundwater is possible given the absence of cement.	Note that this waste stream has been identified by Sellafield Ltd as having an incorrect fingerprint in the National Inventory (Khan <i>et al.</i> , 2009).

Waste Stream	% of ¹⁴ C in LLWR	Waste Stream Description	Waste Form (as given in 2007 National Inventory)	¹⁴ C release potential to gas	¹⁴ C release potential to GW	NNL Comment on materials present within the waste stream	NNL Comment on Inventory and Upper Bound Uncertainty of the waste stream
2C13	1.4	Large Items from Reactor Areas	Not stated	Medium	Medium	Potential exists for ¹⁴ CH ₄ and ¹⁴ CO ₂ release during the corrosion of activated metals (if this is the source of the ¹⁴ C). Although cellulose is present, its nature (wood, large items) lowers the potential for gas generation. The release of ¹⁴ C to groundwater is possible given the absence of cement.	This waste stream has an arising upper band of A and a lower band of D, showing that there is a skew in confidence by the compiler limiting the potential for the ¹⁴ C to go much higher, but having the potential to fall by 2 orders of magnitude.
9C911	1.2	Care & Maintenance Preparation : Reactor and Boiler Systems LLW	The chemical form of ¹⁴ C has not been determined but may be graphite.	Medium	Medium	Significant organic content could generate CH ₄ gas and generate acidity promoting leaching to groundwater. The release of ¹⁴ C to groundwater is possible given the absence of cement.	This waste stream does not arise in the National Inventory until 2009; therefore the basis of the estimated activity during compilation is a derived estimate and not based on measured values. There is an order of magnitude bounding to the ¹⁴ C activity associated with this waste stream.
2D109	1.1	Miscellaneous Plants Initial/Interim Decommissioning: Processing Plants, Tanks, Silos etc.	Not stated	Medium	Medium	Potential exists for ¹⁴ CH ₄ and ¹⁴ CO ₂ release during the corrosion of activated metals. There is the potential for gas generation from organics. The release of ¹⁴ C to groundwater is possible given the absence of cement.	There is a large upper bound value (C) associated with this waste stream, which if applied could increase the total amount of ¹⁴ C in the future vaults by 10%.
2X31	1.1	Oxide Ponds: THORP Flask Maintenance LLW	Not stated	Medium	Medium	Potential exists for ¹⁴ CH ₄ and ¹⁴ CO ₂ release during the corrosion of activated metals. There is the potential for gas generation from organics. The release of ¹⁴ C to groundwater is possible given the absence of cement.	Note that this waste stream has been identified by Sellafield Ltd as having an incorrect fingerprint in the National Inventory (Khan <i>et al.</i> , 2009).
7D26/C	1.1	Conditioned Low Level Ion Exchange Resin (excl. Plant Decontamination)	¹⁴ C is present on resin in a number of different chemical forms, mainly carbonate, organic acids and carbide in magnetite.	High	Medium	As for 7D39/C (submarine resins), the evidence of carbide in magnetite should be examined.	This waste stream has been consigned to Vault 8 in the past and has a confirmed ¹⁴ C content. The compiler has assigned an A Band to the upper uncertainty which projects a good confidence in the estimate.
2C921	1.0	Care and Maintenance Preparation Ponds LLW	The chemical form of ¹⁴ C has not been determined.	Low	Low	The potential for gas generation is low. In addition, the presence of cement may immobilise carbonate.	This waste stream is stated to have stocks and arisings, but the ¹⁴ C activity is estimated, not measured. There is an order of magnitude bounding to the declared activity concentration.
6H02	1.0	LLW (Minor Users)	Not stated	Low	Low	The potential for gas generation is low. In addition, the presence of cement may immobilise carbonate.	The activity is based on the mean annual activity disposed to Vault 8 between 2001 and 2004. There is an uncertainty that waste generated over this time period may not represent wastes generated in the future.

Waste Stream	% of ¹⁴ C in LLWR	Waste Stream Description	Waste Form (as given in 2007 National Inventory)	¹⁴ C release potential to gas	¹⁴ C release potential to GW	NNL Comment on materials present within the waste stream	NNL Comment on Inventory and Upper Bound Uncertainty of the waste stream
1B03	0.7	LLW Non-Compactable Non-Drummable	~80% of waste may degrade to produce gases.	Low	Low	Stainless steel would have a low corrosion rate and may not generate significant gas. Hydrogen is likely to diffuse rapidly and be consumed by biogeochemical processes. The evidence for gas production should be examined.	There is a reasonable confidence associated with this waste stream
1B02	0.1	Non-compactable non-drummable	Not stated	Not Assessed	Not Assessed	The materials of this waste stream have not been assessed for groundwater or gaseous release.	1B02 from GE Healthcare Cardiff has been present in consignments to Vault 8 which, if assume an activity split based on the volumetric ratios with the other streams present, represents about 34GBq of ¹⁴ C in about 940m ³ (0.04 GBq/m ³). The forecast is 4 GBq ¹⁴ C in 33 m ³ (0.1 GBq/m ³).
7E23	0.1	Metallic Waste	Trace quantities of ¹⁴ C are present in metal salts e.g. carbonates.	Not Assessed	Not Assessed	The materials of this waste stream have not been assessed for groundwater or gaseous release.	This is a metallic waste from Rosyth. Its future forecast of ¹⁴ C is 2 GBq in 238 m ³ of waste (0.008 GBq/m ³). It has consigned 20 GBq of ¹⁴ C to the LLWR in 615 m ³ (0.03 GBq/m ³). This apparent drop in concentration should be examined.
2N05/C	N/A	Vault 8 disposals	Not stated	Not Assessed	Not Assessed	The materials of this waste stream have not been assessed for groundwater or gaseous release.	This waste stream has been included as a stock in the UK National Inventory and represents the contents of Vault 8.
1A02	0	Non-compactable drummable	Not stated	Not Assessed	Not Assessed	The materials of this waste stream have not been assessed for groundwater or gaseous release.	This waste stream has consigned ¹⁴ C in the past to Vault 8 but its ¹⁴ C content is classed as not significant in the National Inventory.
1A03	0	Non-compactable non-drummable	Not stated	Not Assessed	Not Assessed	The materials of this waste stream have not been assessed for groundwater or gaseous release.	This waste stream has consigned ¹⁴ C in the past to Vault 8 but its ¹⁴ C content is classed as not significant in the national inventory.
7A116	0	Decommissioning LLW suitable for disposal at the LLWR - miscellaneous	Not stated	Not Assessed	Not Assessed	The materials of this waste stream have not been assessed for groundwater or gaseous release.	This waste stream contains unaccounted for activity (difference between total activity and sum of radionuclide activity). If a proportion of this is ¹⁴ C, then it could be significant.

4.3. Assessment of material/activity association in ^{14}C -bearing wastes

The physical form of the disposed waste can have a significant influence on the timing and nature of contaminant release to the biosphere and geosphere. For example, degradation of ^{14}C -bearing organics may lead to release in the near-term of ^{14}C -labelled gases, whilst ^{14}C in the form of activated graphite blocks would be likely to have long-term stability. It is therefore important to understand how the ^{14}C activity is distributed in relation to the material content of the waste.

Whether the activity is present as contamination or as activated material is of relevance in the safety assessment. ^{14}C may be present as surface contamination, in which case it could easily be leached, whilst activation products will be present within solid phases and will be less-readily leached.

Ball *et al.*, (2008) considered that all ^{14}C activity was associated solely with the cellulose content of the waste. Clearly, some waste streams contain ^{14}C and have no, or very little, cellulosic content and therefore this assumption is considered pessimistic.

In addition, ^{14}C activity may potentially be associated with only one specific material type within a waste stream. This is an important point to consider, as activity concentrations stated in the 2007 National Inventory apply as an average across the whole waste stream. If it could be determined that ^{14}C activity is associated with only a fraction of a waste stream, then the actual concentration of ^{14}C within that fraction would be higher than stated for the total waste stream in the 2007 National Inventory. For example, consider a decommissioning waste stream with 50% graphite, 25% stainless steel and 25% soil by volume and a ^{14}C concentration of $1.0\text{E}-05\text{ TBq/m}^3$. If it were subsequently determined that the ^{14}C was contained entirely within the graphite component, which represents only 50% of the waste volume, then the actual concentration of ^{14}C within the graphite would be $2.0\text{E}-05\text{ TBq/m}^3$; twice that of the whole waste stream.

To investigate these issues, an assessment of the 2007 National Inventory data was undertaken to identify the material content of the ^{14}C -bearing wastes. ^{14}C activities were then apportioned for each waste stream in two distinct ways:

- By equal distribution across the waste stream using the 2007 National Inventory ^{14}C concentrations; and
- By allocation in total separately to each material in each waste stream, to determine the concentration if ^{14}C were associated with one material type: a theoretical maximum.

The calculated ^{14}C concentrations by material were then grouped according to 14 concentration bands in descending order of magnitude from greater than 10 TBq/m^3 to zero, as shown in Table 8.

Table 8 Concentration bandings used in material ^{14}C activity assessment

Band	1	2	3	4	5	6	7	8	9	10	11	12	13	14
From (TBq/m^3)	>1.00E+01	1.0E+01	1.0E+00	1.0E-01	1.0E-02	1.0E-03	1.0E-04	1.0E-05	1.0E-06	1.0E-07	1.0E-08	1.0E-09	1.0E-10	0.0E+00
To (TBq/m^3)	1.0E+01	1.0E+00	1.0E-01	1.0E-02	1.0E-03	1.0E-04	1.0E-05	1.0E-06	1.0E-07	1.0E-08	1.0E-09	1.0E-10	1.0E-17	0.0E+00

Activities and volumes were assessed separately for wastes with ^{14}C activity present as contamination or as activated material, to enable an examination of the relative impacts.

For waste streams with ^{14}C present as contamination, the results of the ^{14}C activity banding by bulk material volume are shown for the material-specific and waste stream average distributions in Table 9 and Table 10 respectively.

For waste streams with ^{14}C present as activated material, the results of the ^{14}C activity banding by bulk material volume are shown for the material-specific and waste stream average distributions in Table 11 and Table 12 respectively.

Further assessment of the results shown in Table 9 to Table 12 was undertaken to identify bands of significant, insignificant and no ^{14}C activity. A particular banding was taken to have significant ^{14}C activity if it represented 1% or more of the total ^{14}C activity for that material group. Bands with non-zero levels of ^{14}C activity less than 1% of the total were taken to be insignificant.

In each of the four tables, significant ^{14}C activities are highlighted in orange, insignificant ^{14}C activities are highlighted in yellow and bands with no ^{14}C activity remain un-highlighted.

It can be seen in Table 9 that the bulk of the volume of Cellulosics and Metals with ^{14}C contamination, where waste stream activity is distributed only to those materials, is in bands 9 and 10 ($1.0\text{E}-06$ to $1.0\text{E}-08$ TBq/m^3); i.e. of low significance. However, the large volumes associated with these materials may increase the likelihood of ^{14}C -labelled gas release. It is noted that around a quarter of contaminated Cellulosics and almost a half of contaminated metal have no associated ^{14}C activity.

The material group having ^{14}C activity in the highest band (band 1) for contaminated wastes, if waste stream ^{14}C activity were associated only to that material type, is Complexants (Table 9). This is due to Complexants being generally a small component of waste streams, representing less than 0.05% of the total waste volume. Similar results are obtained for Liquids and Graphite, which also each represent less than 0.05% of the total waste volume. Whilst ^{14}C concentrations could, in theory, be high in these materials, the associated volumes are very small and therefore unlikely to be of significance in relation to ^{14}C release.

Table 10 shows the waste stream average distribution of ^{14}C activity in contaminated materials. This is considered to represent the best estimate distribution of ^{14}C activity across the materials, rather than the theoretical maximum shown in Table 9. It is seen that concentrations of ^{14}C are generally lower than those for the equivalent material-specific distributions shown in Table 9. This is expected, since materials are assigned a proportional share of the waste stream activity rather than each receiving all activity in the waste stream.

The bulk of the volume associated with ^{14}C -contaminated Cellulosics is focused in band 10 in Table 10. Volumes associated with ^{14}C -contaminated Metals are, as for Table 9, predominantly in bands 9 and 10, but with reduced amounts and a significant increase in band 11.

It can be seen that the highest activity band exhibited for all material types in Table 10 is band 5; a reduction of around four orders of magnitude.

Table 11 shows the distribution of volume by ^{14}C concentration for activated wastes, apportioned by material type. Activated wastes, which arise predominantly from reactor decommissioning, account for only around 16% of total arisings by volume. Around 70% of these wastes are Metals, whilst Cellulosics contribute only around 2%.

It is shown in Table 11 that around half of the activated Metals volume associated with ^{14}C is in band 8 ($1.0\text{E}-05$ to $1.0\text{E}-06$ TBq/m^3); around one to two orders of magnitude higher than for ^{14}C -contaminated Metals (Table 9). This is potentially significant, particularly since the activity within activated metals cannot be removed through decontamination prior to disposal and may subsequently be released through corrosion in the vault.

It is of interest to note that the best estimate, proportional distribution of ^{14}C activity in activated Metals presented in Table 12 also has around half the volume in band 8. This is because the waste streams contributing the majority of activity are almost 100%

activated metal, thereby giving similar results whether activity is allocated in its entirety, or is allocated proportionally, to each material.

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Table 9 Bulk material volumes by ¹⁴C concentration group for contaminated waste, activity distributed by material

Material Type	Total Volume (m ³)	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Cellulosics	82,017	0	0	0.018	0.005	268	433	2,710	2,480	22,700	18,600	1,180	10,100	1,040	22,500
Complexants	312	0.001	0.169	0.295	7.67	1.95	0.47	9.5	275	0.06	0.0022	0	0	0	16
Graphite	305	0	0.01	0.514	0.045	0.69	11.3	180	0.47	0.3	0	0	0	0	111
Inorganic	13,655	0	0	0.066	0.051	14.1	1,930	943	1,180	2,410	690	108	4	40	6,340
Liquid	314	0	0.002	0.297	0.294	0.208	156	0.197	0.15	0.12	0.002	3.2	0	0	154
Metals	285,459	0	0	0.202	21.7	8.59	2,760	4,350	8,680	75,900	46,800	8,950	9,840	3,900	124,000
Organic	2,702	0	0	1.210	22.9	17	656	402	484	91	0	695	0	0	334
Plastic / Rubber	84,812	0	0	0.049	0.036	5.41	729	3,800	11,300	25,300	11,200	1,110	3,350	386	27,700
Soil / Rubble	160,666	0	0	0	0.001	343	522	1,320	23,500	8,370	11,200	59,000	180	20,200	36,000
Unknown	11,189	0	0	0	0	0	2.22	0	245	593	30	0	0	0	10,300
All Materials	641,432	0	0	3	53	659	7,200	13,715	48,145	135,365	88,520	71,046	23,474	25,566	227,455
		0%	0%	0%	0%	0%	1%	2%	8%	21%	14%	11%	4%	4%	35%

Table 10 Bulk material volumes by ¹⁴C concentration group for contaminated waste, activity distributed by waste stream

Material Type	Total Volume (m ³)	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Cellulosics	82,017	0	0	0	0	0	6.08	643	2,708	11,371	27,429	6,124	8,701	2,497	22,538
Complexants	312	0	0	0	0	0.16	7.6	0.08	1.3	1.1	284	0.21	0.67	0.06	16
Graphite	305	0	0	0	0	0	0	0.52	190	0.28	3.1	0.01	0.24	0.06	111
Inorganic	13,655	0	0	0	0	0	18	1,984	596	752	3,318	302	190	152	6,344
Liquid	314	0	0	0	0	0	154	2.52	0.63	0.06	0.03	3	0.15	0.12	154
Metals	285,459	0	0	0	0	22	37	3,564	8,899	65,111	36,262	32,921	10,287	4,091	124,266
Organic	2,702	0	0	0	0	30	537	56	129	340	478	105	628	66	334
Plastic / Rubber	84,812	0	0	0	0	0	5.99	940	6,156	15,969	22,246	7,970	2,543	1,274	27,707
Soil / Rubble	160,666	0	0	0	0	65	117	1,025	19,153	7,070	17,442	59,364	0.07	20,463	35,967
Unknown	11,189	0	0	0	0	0	0	2.22	209	417	213	29	0	0	10,319
All Materials	641,432	0	0	0	0	117	882	8,217	38,041	101,032	107,675	106,818	22,350	28,543	227,757
		0%	0%	0%	0%	0%	0%	1%	6%	16%	17%	17%	3%	4%	36%

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Table 11 Bulk material volumes by ¹⁴C concentration group for activated waste, activity distributed by material

Material Type	Total Volume (m ³)	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Cellulosics	2,927	0	0	0	0	0	576	1,330	598	410	0	0.52	0	15	0
Complexants	30	0	0	0	0.08	1.43	0	0	0.19	0	0	0	0	0	29
Graphite	3.8	0	0	0	2.23	0	0	1.6	0	0	0	0	0	0	0
Inorganic	3,852	0	0	0.67	0	55.1	440	414	14	0	0	0	5	0	2,920
Liquid	0.5	0	0	0.005	0	0	0	0	0	0	0	0	0	0	1
Metals	87,213	0	0	0	0	0	438	4,270	46,500	11,600	2,280	0	5.3	22	22,100
Organic	1,018	0	0	0.551	0	0.013	0	0	0.19	987	0.026	0.24	0	0	30
Plastic / Rubber	3,617	0	0	0	0	20.5	87	1,130	663	1,570	0	1.05	0	0	145
Soil / Rubble	22,128	0	0	0	4.46	0	14	4,600	5,290	8	9,260	0	0	451	2,500
Unknown	1,453	0	0	0	0	0	0	12	0	0	0	0	0	0	1,440
All Materials	122,242	0	0	1	7	77	1,555	11,757	53,065	14,575	11,540	2	10	487	29,165
		0%	0%	0%	0%	0%	1%	10%	43%	12%	9%	0%	0%	0%	24%

Table 12 Bulk material volumes by ¹⁴C concentration group for activated waste, activity distributed by waste stream

Material Type	Total Volume (m ³)	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Cellulosics	2,927	0	0	0	0	0	0	520	1,839	143	410	0	0	15	0
Complexants	30	0	0	0	0	0	0	0	1.5	0.01	0.19	0	0	0	29
Graphite	3.8	0	0	0	0	0	0	2.23	2	0	0	0	0	0	0
Inorganic	3,852	0	0	0	0	0	0	56	848	20	0.19	0	0	5	2,924
Liquid	0.5	0	0	0	0	0	0	0	0	0	0	0	0	0	1
Metals	87,213	0	0	0	0	0	0	4,490	46,499	10,459	3,615	0	0	27	22,123
Organic	1,018	0	0	0	0	0	0	0.6	0	0	987	0	0	0.27	30
Plastic / Rubber	3,617	0	0	0	0	0	0	78	1,589	234	1,570	0	0	1.05	145
Soil / Rubble	22,128	0	0	0	0	0	0	4,593	5,308	10	9,269	0	0	451	2,497
Unknown	1,453	0	0	0	0	0	0	11.50	0	0	0	0	0	0	1,441
All Materials	122,242	0	0	0	0	0	0	9,751	56,086	10,866	15,851	0	0	500	29,189
		0%	0%	0%	0%	0%	0%	8%	46%	9%	13%	0%	0%	0%	24%

Figure 3 shows the volumetric distribution of wastes containing significant, insignificant and no ^{14}C activity over time for Metals.

It is shown that wastes containing significant ^{14}C activity arise up to around 2060, with a significant 'spike' of arisings of around 7,000 m^3 per year between 2009 and 2017. Tables 4 to 7 have shown that around 70% of future metal arisings having associated ^{14}C activity are surface-contaminated rather than activated. In addition, activated metals from reactor decommissioning are generally forecast to arise in later years, therefore it can be expected that the large majority of metallic wastes containing ^{14}C activity arising in the immediate future will be surface-contaminated. The early development and deployment of treatment technologies to segregate and decontaminate metallic wastes prior to LLWR disposal is therefore imperative to reduce impact from ^{14}C .

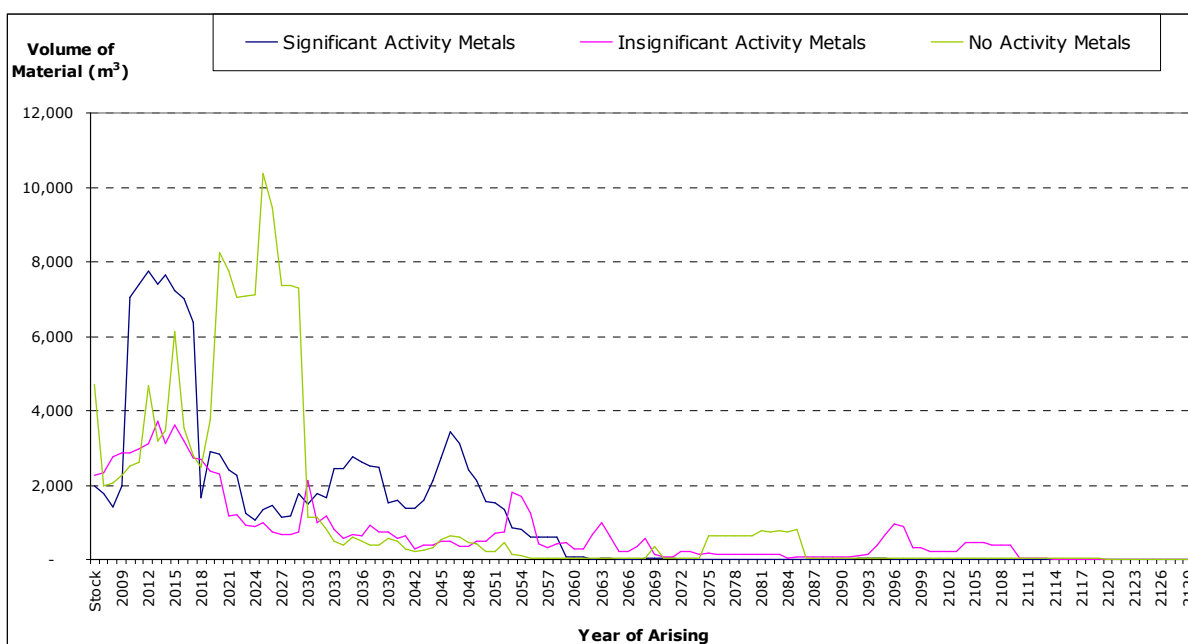


Figure 3 Forecast ^{14}C activity distribution by volume in UK LLW Metals

Figure 4 shows the volumetric distribution of wastes containing significant, insignificant and no ^{14}C activity over time for Cellulosics.

It is shown that the highest rate of future Cellulosics arisings containing significant ^{14}C is forecast to occur over the next few years, with a peak of just under 1,800 m^3/yr by around 2011. Arisings are then predicted to reduce significantly, decreasing to around 600 m^3/yr by 2027, then down to below 200 m^3/yr by 2060. This is in line with expectations, as soft operational wastes such as paper and cotton are replaced with time by structural materials from plant decommissioning.

The findings indicate therefore that the impact from ^{14}C -bearing gas generation from the degradation of cellulose waste within the LLWR future vaults is likely to be greatest from wastes disposed in the early vaults (i.e. Vaults 9 to 11). Options for the treatment of cellulosics and other degradable wastes prior to disposal, such as incineration, should be developed and implemented as soon as possible to reduce the potential impact from the release of ^{14}C -bearing gas.

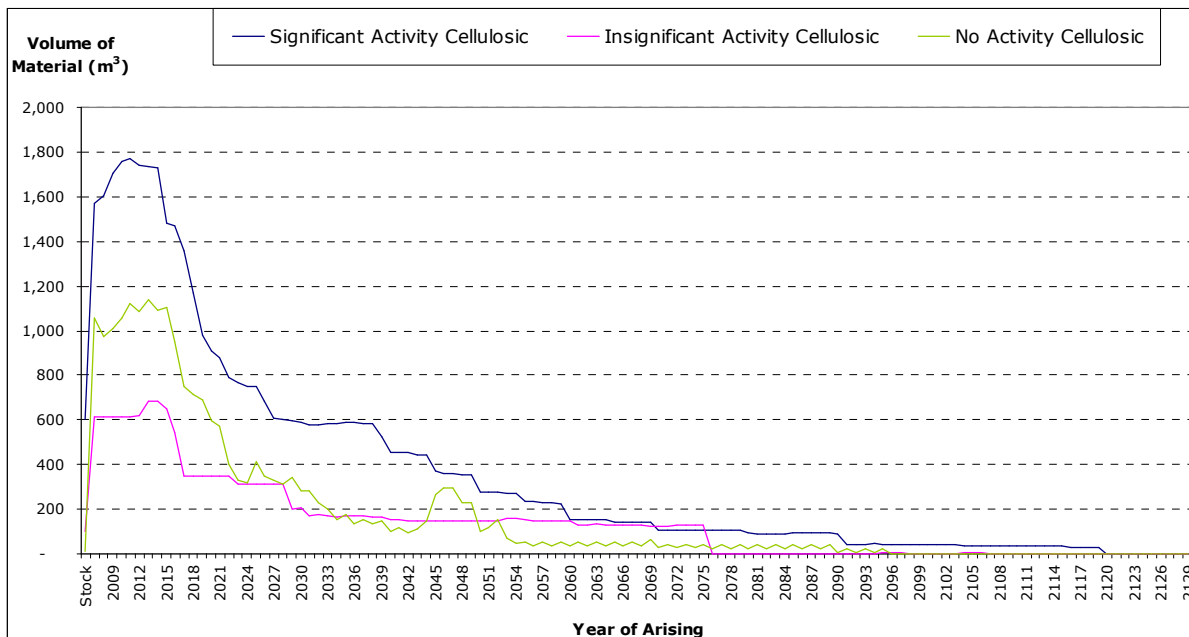


Figure 4 Forecast ¹⁴C activity distribution by volume in UK LLW Cellulosics

The potential impact of implementing alternative treatment technologies for cellulosic and metallic wastes prior to disposal at the LLWR is examined further in Section 5.

4.4. Additional information gathering

The gathering of additional data to reduce uncertainty in the future ¹⁴C inventory fell into three categories:

- WSCDs for waste streams currently consigning to the LLWR;
- Direct discussion with waste producers for waste streams not yet arising; and
- Assessment of Waste Receipt Monitoring data from Vault 8 disposals.

Table 13 Shows the applicable data sources for the waste streams identified for further information to be sought, as listed in Table 7, and summarises the key findings.

Table 13 Sources identified for further information on key ¹⁴C waste streams

Waste Stream	Further information gathered from	Key findings
7D39/C	Devonport consignor	The inventory is a derived best estimate. A process (ModuloX™) is under development to remove ¹⁴ C from the waste, following which, if successful, the waste will contain little or no ¹⁴ C. If the process is not successful and the ¹⁴ C cannot be removed, then the waste will not be deemed suitable for LLWR disposal and an alternative disposal route will be sought. Either way, the impact of ¹⁴ C from this waste stream on LLWR future disposals is expected to be minimal.

Waste Stream	Further information gathered from	Key findings
9E958	Oldbury consignor	<p>The consignor stated that the radionuclide fingerprint for 9E958 is based on that for the waste stream 9E58. However, it was noted that the volume for 9E58 is only 61.2 m³. The 2007 National Inventory states for 9E58 that ¹⁴C activity arises from graphite dust contamination. That it is reasonable to extrapolate from 9E58 to 9E958 with a volume of around 5,000 m³ was questioned with the consignor.</p> <p>The consignor later confirmed that the volume for 9E958 given in the 2007 National Inventory is approximately 4,000 m³, but confidence in the fingerprint was low.</p> <p>Further contact was made with the consignor, who explained that a programme to assess the VLLW content of Magnox wastes would be undertaken within the coming year, which could result in certain portions of future arisings from waste streams such as 9E958 being designated as VLLW.</p>
5G303	Winfrith consignor	<p>The consignor confirmed that the ¹⁴C activity stated in the 2007 National Inventory is an upper estimate. The consignor confirmed that the ¹⁴C activity is associated predominantly with the graphite core, which represents a small fraction of the waste; therefore the majority of the waste may have little or no ¹⁴C. The 2007 National Inventory does not provide a split of volumes for core and non-core components. The consignor did not have this split to hand, but offered to investigate further. No further information has been provided at the time of writing.</p>
3S06	Current WSCD	<p>Assessment of the WSCD for this waste stream shows the total activity of ¹⁴C to be around 22.6 GBq in a volume of 1,054 m³. However, the 2007 National Inventory states a total activity of 110 GBq in a volume of 500 m³; a concentration of around 10 times greater. Discussion with the consignor is required to understand these differences.</p>
5F307	Windscale consignor	<p>The consignor confirmed that the waste stream 5F307 is made up from four sub-streams, of which only one, 2X307/4, contains ¹⁴C.</p>
2X927	Current WSCD	<p>The consignor advised that a review of Sellafield LLW radionuclide fingerprints was being undertaken, following the discovery of a number of significant overestimations at around three orders of magnitude. For 2X927, the fingerprints given in the 2007 National Inventory were based on the WSCD for this waste stream; however the data had been transposed with incorrect units. Further information is provided in Khan <i>et al.</i>, 2009.</p>
2C920	Chapelcross consignor	<p>The consignor stated that the radionuclide fingerprint for 2C920 was based on that for the operational waste stream 2C11. The radionuclide fingerprints for both 2C920 and 2C11 in the 2007 National Inventory were compared with that in the current WSCD for 2C11. The National Inventory activities for 2C11 were found to be between 10 and 100 times greater than those given in the WSCD for this waste stream. The findings were queried with the consignor by email, but no further information has been provided at the time of writing.</p>
7E26	Current WSCD	<p>A current WSCD was not supplied for this waste stream.</p>
2X21	Current WSCD	<p>As for 2X927, the fingerprints given in the 2007 National Inventory were based on the WSCD for this waste stream; however the data had been transposed with incorrect units to give an overestimate of around three orders of magnitude. Further information is provided in Khan <i>et al.</i>, 2009.</p>
2C13	Current WSCD	<p>The WSCD showed fingerprint activities to be approximately 100 times lower than stated in the 2007 National Inventory. Further investigation is required to understand the reasons for this difference.</p>
9C911	Dungeness A consignor	<p>Contact could not be made with the consignor at the time of writing.</p>

Waste Stream	Further information gathered from	Key findings
2D109	Sellafield consignor	The consignor advised that the future forecast radionuclide fingerprint for this waste stream had been derived from actual past disposal records for the plants it represented in 2001. However, it is not expected that the mix of plants and activities that contribute to this stream will be the same in the future; therefore there is low confidence that the radionuclide fingerprint ratios and concentrations given in the 2007 National Inventory are representative. No improved data are available at the time of writing, therefore the uncertainty bands given in the National Inventory should apply until more up-to-date information is provided.
2X31	Current WSCD	As for 2X927, the fingerprints given in the 2007 National Inventory were based on the WSCD for this waste stream; however the data had been transposed with incorrect units to give an overestimate of around three orders of magnitude. Further information is provided in Khan <i>et al.</i> (2009).
7D26/C	Devonport consignor	The consignor confirmed that the ^{14}C activity is a measured value, and is present in the waste in the form of carbonate, organic acids and carbide (magnetite). It is understood that LLWR have refused D5 consignments of this waste stream in the past due to the ^{14}C content.
2C921	Chapelcross consignor	The consignor stated that the radionuclide fingerprint for 2C921 was based on that for the operational waste stream 2C10, although further investigation revealed that it was based on 2C14. The radionuclide fingerprints for both 2C921 and 2C14 in the 2007 National Inventory were compared with that in the current WSCD for 2C14. The National Inventory activities for 2C14 were found to be around 2.5 times greater than those given in the WSCD for this waste stream. In addition, the activities in the fingerprint for 2C921 were found to be around 5 times less than those for 2C14. The findings were queried with the consignor by email, but no further information has been provided at the time of writing.
6H02	No source identified	No contact was provided for this waste stream.
1B03	WSCD (expired Dec 08)	There is no current WSCD for this waste stream, the most recent one (2005) having expired in December 2008. A check was made of two WSCDs, dated 2000 and 2005. It was found that the 2000 WSCD had a ^{14}C activity of 15 MBq/te, whilst the 2005 WSCD showed a ^{14}C activity of only 0.02 MBq/te. Assessment of the historical disposal records shows significantly higher actual activities disposed for this waste stream. Further investigation is required to understand the reasons for these differences.
1B02	WSCD (expired Dec 08)	There is no current WSCD for this waste stream, the most recent one (2005) having expired in December 2008. A check was made of the two previous WSCDs, dated 2000 and 2005. It was found that the 2000 WSCD had a ^{14}C activity of 115 MBq/te, whilst the 2005 WSCD showed a ^{14}C activity of only 0.1 MBq/te. Assessment of the historical disposal records shows significantly higher actual activities disposed for this waste stream. Further investigation is required to understand the reasons for these differences.
1A02	WSCD (expired Dec 08)	There is no current WSCD for this waste stream, the most recent one (2005) having expired in December 2008. A check was made of the two previous WSCDs, dated 1997 and 2005. It was found that the ^{14}C activity in the 1997 WSCD was around 11.6% of the total activity, but this dropped to only 0.1% of the total activity in the 2005 WSCD. Assessment of the historical disposal records shows significantly higher actual activities disposed for this waste stream. Further investigation is required to understand the reasons for these differences.
1A03	WSCD (expired Dec 08)	There is no current WSCD for this waste stream, the most recent one (2005) having expired in December 2008. A check was made of the two previous WSCDs, dated 1997 and 2005. It was found that the ^{14}C activity in the 1997 WSCD was around 0.2% of the total activity, but this halved to 0.1% of the total activity in the 2005 WSCD. Assessment of the historical disposal records shows significantly higher actual activities disposed for this waste stream. Further investigation is required to understand the reasons for these differences.
7E23	Current WSCD	A current WSCD was not supplied for this waste stream.

Waste Stream	Further information gathered from	Key findings
2N05/C	No source identified	No contact was provided for this waste stream.
7A116	Current WSCD	A current WSCD was not supplied for this waste stream.

WRM data were obtained for disposals to Vault 8 between 2002 and 2008 and assessed for the waste streams identified in Table 7. The WRM programme actively measures the activity content of consignments for comparison with the declared activity on the disposal records. ^{14}C cannot be accurately measured by the High Resolution Gamma Scanner used in the WRM programme, but it was considered that the observed results for other radionuclides could be used to determine the overall accuracy of the inventory data, and hence infer the accuracy of the ^{14}C inventory.

Consignments containing single waste streams are of particular interest, as obtained results are applicable only to that waste stream. Where consignments contain a mixture of waste streams, it is often not possible to determine the relative contributions from the different components. It was found that monitoring had been undertaken for consignments containing only 5F307, 2X31 and 2X927. For these waste streams, the following observations were made (Table 14).

Table 14 Comments in Waste Receipt Monitoring Records for ^{14}C -bearing waste

Waste Stream	Date of assessment	Comment
5F307	23/12/2002	Failed WRAP by overestimating ^{60}Co
	03/10/2006	Accurate declaration. Slight overestimate on ^{137}Cs
2X31	27/07/2005	No comment available for this assessment. Mixed consignment containing 2X31 overestimated ^{137}Co , ^{60}Co , ^{125}Sb and ^{106}Ru on
2X927	15/02/2006	No comment available for this assessment
	17/01/2007	Pass. Low activity. Over-declared on major gammas
	20/12/2007	Low activity. WRAP pass

The Sizewell B waste stream 3S07 gave good agreement against the D4 and the WSCD in the WRM, but differences between these and the 2007 National Inventory fingerprints were found for this waste stream.

The general trend in WRM is one of over-declaration of radionuclide activities in the disposal forms. It is observed that Sellafield waste streams in particular tend to be over-declared, whilst greater accuracy is demonstrated for non-Sellafield wastes.

4.5. Discussion of findings

The findings of the assessment of underpinning inventory data for the key waste streams have shown a general trend of overestimation in the 2007 National Inventory. Assessment of WSCDs and discussion with the consignors has revealed that inventories for the few waste streams representing around 75% of future forecast ^{14}C activity may be revised. The top contributing waste stream, 7D39/C, is being assessed for suitability in applying a treatment process to remove the large majority of ^{14}C activity. Up to around 25% of future forecast ^{14}C for LLWR disposal may be removed if successful.

The information provided by the waste producers has been qualitative and has not enabled bounding calculations to be performed. A Probability Density Function (PDF) for each waste stream would be required to both understand the basis of the best estimate

provided within the 2007 UK National Inventory and to produce an overall PDF for ^{14}C . Table 15 presents the information discovered to date and shows the basis of the confidence in the prediction for each waste stream.

Table 15 Results of the assessment of the ^{14}C inventory for key contributing waste streams

Stream	Comment	National Inventory Estimate (GBq)
7D39/C	The anticipated ModulOx™ treatment process will result in the removal of a significant proportion of the ^{14}C activity present in the ion exchange resin, which is currently at ILW levels as waste stream 7D41. At this currently declared level there is the real possibility that the consignments will not conform to the LLWR's Conditions for Acceptance (CFA). Devonport have experienced D5 preventions for operational streams such as 7D26/C. Therefore the anticipated ^{14}C activity of 442 GBq associated with this stream would be significantly lower should the waste be consigned to the LLWR.	442
9E958	Discussions with Magnox Oldbury have identified a large uncertainty associated with this waste stream's radionuclide fingerprint. Whilst there is a robust case for the stated volume estimated to arise, there is a good likelihood that the radionuclide concentrations are an overestimate, as they are based on another stream with a much smaller volume. It is likely that decommissioning waste characterisation work planned to be undertaken by Magnox North over the next few years will identify a large proportion of the volumes of their decommissioning waste streams as Very Low Level Waste (VLLW) and the fingerprint will only be applicable to the remaining volume.	355
5G303	Discussions with Winfrith personnel have identified that the ^{14}C activity stated in the 2007 National Inventory is an upper estimate. Activity is associated predominantly with the graphite core, which represents a small fraction of the waste; therefore the majority of the waste may have little or no ^{14}C .	119
3S06	No contact was made with the waste producer for 3S06. An assessment of the WSCD and previous disposals to Vault 8 have shown that the WSCD declares a lifetime activity for ^{14}C of 22 GBq. It is unlikely therefore that the value of 111 GBq declared in the UK National Inventory will be exceeded.	111
5F307	The ^{14}C concentration was incorrectly based on the fingerprint of a sub-stream and did not take into account the drop in concentration that would occur when taking the other sub-streams into account when constructing the 5F307 composite. The majority of the waste contains no significant ^{14}C , resulting in total ^{14}C of no more than 1 GBq.	105
2C920	The ^{14}C concentration is based on the operational stream 2C11. The volume is much larger for this C&M Preps stream and it could be expected that the overall ^{14}C concentration should drop as a result.	86
7E26	No additional information was reviewed for this stream. However, the waste stream 7E26 has been consigned to Vault 8.	33
2C13	The WSCD showed the fingerprint activities to be approximately 100 times lower than stated in the 2007 UK National Inventory. No discussions were made with the waste producer.	24
9C911	No additional information was reviewed for this waste stream.	22
2D109	This stream fingerprint has not changed since the 2001 UK National Inventory. It is a composite waste stream constructed to represent wastes arisings from the decommissioning of a number of building on the Sellafield site. Discussion with the waste producer have identified that some of the buildings may have changed. . It should be noted that the volumes are large (67301 m ³), so a small change in calculated concentration could make the ^{14}C inventory of this waste stream higher or lower than declared	20

Stream	Comment	National Inventory Estimate (GBq)
7D26/C	Discussion with the waste producer and a review of the disposal records have shown that 7D26 has been consigned to Vault 8 at the LLWR. There was also anecdotal evidence from the waste producer that a variation to the waste stream (a form D5) has been refused for this stream. This waste is unlikely to arrive at the LLWR unless changes to the CFA occur.	19
2C921	Similar to 2C920, this decommissioning stream takes its fingerprint from an operational stream with a much lower volume.	18
6H02	This stream is based on previous disposals to the LLWR and will have included MOD and Amersham disposals. Potential for double counting of ^{14}C from these sources.	17
Streams with <1% ^{14}C present	There are a number of streams containing ^{14}C activity but each does not contain >1% of the total ^{14}C activity. Applicability to apply the upper band for all streams has a decreased likelihood.	241

* Magnox will be undertaking a programme of work in 2009 to scope a study to determine the quantity of VLLW present in Decommissioning wastes.

It is recommended that this is monitored closely by the LLWR, so that such information can be expediently used in good time for the ESC in 2011.

The ^{14}C in the trenches is attributable to disposals predominantly from outwith the nuclear industry. Activated products in Vault 8 have typically arisen from the servicing of the UK's nuclear submarine fleet. Some of these have been consigned without a UK National Inventory identifier (an LLWR allocated waste stream number has been used) and have been used to represent consignments from Defence sites in general. Vault 8 also contains some ^{14}C from the decommissioning of reactors, such as the universities research reactor (URR), Risley and the Windscale AGR. The URR has been included in the small users component to represent ^{14}C disposals to the trenches.

Activated metals and concretes were not typically disposed to the trenches, therefore the evaluation of the ^{14}C content in the trench is likely to be conservative for these types of wastes, since the material composition of the trenches has been based on wastes consigned to Vault 8 and in the 2004 UK National Inventory.

It is known from the waste tracking system database that GE Healthcare Ltd's sites at Amersham and Cardiff have consigned to Vault 8, 38 GBq and 29 GBq of ^{14}C in 1,013 m³ and 5,875 m³ of waste respectively. A review of historic Amersham consignments to the trenches has shown that the ^{14}C content present in the wastes was greater both as a percentage of the total activity and in terms of the total activity in their wastes. Of 441 consignments to the trenches between 1980 and 1987, only 75 consignments (17%) were stated to contain ^{14}C . Upon closer inspection, the average ^{14}C content in the consignment was about 33% of the total activity.

Amersham clearly used to consign more active wastes, and the inventory for both the Vault 8 and the future vaults wastes may not represent this appropriately.

5. Impact of potential alternative treatment and packaging technologies

It is recognised that alternative waste treatment technologies will be required to manage future capacity at the LLWR, such that all forecast arisings may be disposed. Future treatment technologies are expected to focus on particular material types, following appropriate segregation of waste. Volume Reduction Factors (VRFs) have been taken from treatments stated in an IAEA technical document on waste treatment technologies (IAEA, 2006). These factors are not specific to LLW or to the UK and are to provide bounding to the reduction in volumes that could be expected from the processes described. Application of the upper bound VRF results in a larger final volume than application of the lower bound VRF.

¹⁴C-bearing wastes contain either activated or contaminated material. The majority of arisings are contaminated, consisting of a mixture of items including metals, rubble, cellulose, plastics and inorganic material. ¹⁴C-activated wastes are predominantly metal or concrete items from reactor decommissioning. ¹⁴C-labelled CO₂ and CH₄ arise in the main from the degradation of ¹⁴C-contaminated cellulose or from the oxidation of ¹⁴C activated or contaminated metals.

Table 16 shows the raw volumes of future forecast UK LLW split into the following four categories:

- Materials containing ¹⁴C by activation;
- Materials containing ¹⁴C by contamination;
- Materials that do contain ¹⁴C; and
- Materials in which ¹⁴C is present but no activity concentration has been determined.

To derive Table 16, the information available on each of the 635 LLW streams was assessed to enable each stream to be assigned to one of the four categories.

Table 17 shows the equivalent ¹⁴C activities in GBq for the volumes given in Table 16. The assumption made in the calculation is that all ¹⁴C activity is allocated to each bulk material present in the waste stream. The total for each bulk material cannot exceed the total ¹⁴C activity expected to be disposed to the LLWR (1,703 GBq). With this methodology, Cellulose cannot be greater than 800 GBq; about 50% of the total ¹⁴C activity.

Table 18 shows the equivalent ¹⁴C activities in GBq for the volumes given in Table 16. The assumption made in the calculation is that the ¹⁴C activity is allocated on a *pro rata* basis to each bulk material present in the waste stream. On this basis, Cellulose would contain only 58 GBq of ¹⁴C activity; approximately 3% of the total ¹⁴C activity expected to be consigned to the LLWR.

The true amount of ¹⁴C within Cellulose could be even lower than 58 GBq given that 13.8 GBq is associated with streams containing activated materials, decreasing the likelihood that ¹⁴C would be associated with the Cellulose component of those streams.

As stated in Section 2.2, Ball *et al.*, (2008) used a future vaults inventory of 7.35 TBq, coupled with a Cellulose Volume Fraction of 3.75E-02 to derive an initial evolution rate for ¹⁴C-labelled gas of 6.37E+07 Bq y⁻¹. Whilst Table 16 shows a Cellulose Volume Fraction of 1.1E-01, it must be remembered that the Cellulose volume provided in Ball *et al.*, (2008) is based on the final package, and therefore the majority of the Cellulose-bearing waste streams would have undergone a volume reduction comparable to the factors typical of high force compaction. Therefore, the Cellulose Volume Fraction is comparable in Table 16, with Ball *et al.*, (2008). The ¹⁴C associated with Cellulose however, has fallen from 7.35 TBq to at most 800 GBq, but more likely to around tens of

GBq. This represents a direct reduction in the calculated amounts of ^{14}C -labelled gas from Cellulosics of between 1 and 2 orders of magnitude.

^{14}C in Metals were not evaluated in Ball *et al.*, (2008). Assessment of Table 17 and Table 18 shows that it is likely that at least half of the ^{14}C forecast to be consigned to the LLWR can be associated with the Metal component of the waste, and potentially up to around 91%. Note that Table 18 shows an approximately equal split between ^{14}C present in Metal from waste streams containing ^{14}C as activated material and ^{14}C present as a contamination. Evaluation of the behaviour of ^{14}C present in Metal as an activation product or as contamination should be considered in future near field studies.

Table 16 Raw volumes (m³) of future forecast UK LLW split by material group

Material Type	^{14}C Activated waste	^{14}C Contaminated waste	Not Present	^{14}C Present but not determined	Total LLWR
Cellulosics	2,927	59,549	22,251	217	84,944
Complexants	30	296	16	0	342
Graphite	4	194	111	0	309
Inorganic	957	7,312	9,183	55	17,507
Liquid	0	160	154	1	315
Metals	66,638	161,732	129,933	14,370	372,672
Organic	1,017	2,368	324	11	3,720
Plastic / Rubber	3,615	57,220	27,532	62	88,429
Soil / Rubble	20,807	127,839	32,341	1,807	182,794
Unknown	11	871	11,660	100	12,642
Total	96,006	417,541	233,504	16,624	763,674
	13%	55%	31%	2%	

Table 17 ^{14}C activity (GBq) allocated to each material, grouped by bulk material

Material Type	^{14}C Activated waste	^{14}C Contaminated waste	Total LLWR
Cellulosics	231	568	799
Complexants	5	747	753
Graphite	119	396	515
Inorganic	325	510	835
Liquid	1	208	209
Metals	396	1,149	1,545
Organic	106	1,195	1,301
Plastic / Rubber	239	571	810
Soil / Rubble	242	1,019	1,261
Unknown	1	3	4

Table 18 ^{14}C activity (GBq) associated with future forecast UK LLW, distributed by waste stream, split by bulk material group

Material Type	^{14}C Activated waste	^{14}C Contaminated waste	Total LLWR
Cellulosics	13.8	44.3	58
Complexants	0.0	5.2	5
Graphite	0.1	0.4	1
Inorganic	2.7	125.6	128
Liquid	0.0	34.0	34
Metals	279.4	350.3	630
Organic	0.0	299.7	300
Plastic / Rubber	5.0	81.1	86
Soil / Rubble	99.1	361.4	460
Unknown	0.2	0.5	1
Total	400	1,302	1,703

It is recognised that a number of potential alternative treatment technologies may be applied to wastes for future LLWR disposal. However, to determine the impact on ^{14}C activity, the assessment is focused on the metallic and cellulosic waste forms. Two alternative treatments are examined and compared for each of these categories. For metallic wastes, these are high force compaction (HFC) and melting; for cellulosic wastes these are high force compaction and incineration.

Other, more detailed assessments of the impact of treatments and technologies are being undertaken within the Environmental Safety Case, more specific to the LLWR. However, it was considered appropriate to provide some indication of the impact of a small number of currently-available treatments, applying some generic values from IAEA, (2006).

Not all metallic or cellulosic wastes are suitable for HFC, dependent on size, thickness, material type or quantity etc. Wood, whilst being grouped within the bulk material Cellulosics, is not compactable in some forms and therefore 100% of Cellulosics cannot be assumed to be high force compactable. No assessment was made of metal content in direct-to LLWR ISOs against metal content in ISOs consigned for high force compaction. It was therefore assumed that only 10% by volume of metallic arisings and 80% of cellulosic arisings would be routed for HFC.

IAEA, (2006) provided two potential volume reduction factors (VRF) for HFC of LLW with a lower bound VRF of 10, leading to a greater volume reduction and an upper bound VRF of 4.

It was assumed that all metallic waste would be suitable for melting, although it is recognised that for larger items some size reduction would be required beforehand. As for HFC, two potential VRFs would apply: an upper VRF of 5 and a lower VRF of 20.

It was assumed that all cellulosic waste would be suitable for incineration. Two potential VRFs would apply: an upper VRF of 7 and a lower VRF of 100.

In addition to the suitability of the metallic or cellulosic material for treatment, a further consideration is segregation. Whilst all metal may in theory be suitable for melting or all cellulose for incineration, the practical difficulties and cost of separating small amounts of material from the other components in the waste stream may outweigh the benefits. Four alternative scenarios for segregating portions of metallic or cellulosic wastes for subsequent treatment were therefore devised as follows:

1. All metal or cellulose available for segregation;
2. All metal or cellulose available for segregation from streams where the metal or cellulose component is greater than 20% by volume;
3. All metal or cellulose available for segregation from streams where the stream volume is greater than 10,000 m³ and the metal or cellulose component is greater than 1% by volume; and
4. All metal or cellulose available for segregation from streams where the metal or cellulose component is greater than 100 m³ by volume.

Table 19 presents the volumes of metal in future forecast UK LLW following the application of HFC and melting, shown separately for each of the four scenarios described above. The total metal volume consigned to the LLWR in each of the four categories has been further broken down to reflect the knowledge of the ^{14}C association with the metal, i.e. Activated ^{14}C (AC); Contaminated with ^{14}C (CC); Present but Not Determined (PND) or Not Present (NP)

From top to bottom, the table presents the workings of the calculations discussed above to allocate the raw volume between the various treatments and then divide by the upper and lower bound factors for those treatments to obtain an indicative disposal volume.

From left to right, the table presents calculated boundaries for metal volumes based on the confidence in which the metal may be segregated from the other materials within the waste stream. This is based on either large volumes of metal (>100 or >10,000 m³) or large percentages of metal present (>20%) within each waste stream.

Some particular points of interest are as follows:

- The theoretical maximum reduction in the total metal volume for disposal at LLWR following melting at the lower VRF where all metal is suitable for melting is 20-fold, giving a final disposal volume of only 18,634 m³ from a raw volume of 372,672 m³. Of this, approximately 12,000 m³ would contain ¹⁴C activity with a total of 630 GBq, assuming no ¹⁴C loss during thermal treatment.
- The scenario where metal can be segregated with a volume of greater than 100 m³ yields approximately 98% of the metal in the future forecast LLW inventory. Selecting only those waste streams with a total volume of greater than 10,000 m³ and a metal content of greater than 1% yields only around 50% of future forecast metal.
- Whilst HFC of metal does not reduce the total raw volume by a large factor, given that only around 10% of metal is compactable, the total volume of raw metal (372,672 m³) is such that there is potential for significant disposal cost savings in applying HFC where appropriate.

The process of metal melting results in a homogenised mass whereby contamination on the surface of the raw waste may become 'locked' inside the conditioned product. An amount of ¹⁴C activity may be lost from the waste during thermal treatments oxidising carbon to form ¹⁴C-labelled gases during the treatment. However, the benefit arises in terms of the loss of potential for subsequent release of ¹⁴C from the waste to the environment because the contamination will not be accessible to groundwater in the short-term, and there will be a reduction in overall surface area of metal, thereby reducing the potential for corrosion. In addition, the high temperatures required for metal melting may provide some benefit in converting organic ¹⁴C species to inorganic species, thereby reducing the potential for release as ¹⁴C-labelled gases. This may be limited; since it is anticipated that ¹⁴C-activation within metal would be predominantly inorganic.

Table 19 Estimated volume changes for future forecast UK LLW metal when applying HFC or metal melting

	All Metal					All Streams where Metal >20%					All Streams where Stream Volume > 10,000m ³ and Metal > 1%					All Streams where Metal Volume > 100m ³				
	LLWR	AC	CC	PND	NP	LLWR	AC	CC	PND	NP	LLWR	AC	CC	PND	NP	LLWR	AC	CC	PND	NP
Metal volume (m ³)	372,672	66,638	161,732	14,370	129,933	353,115	64,850	151,682	13,864	122,719	195,043	40,515	90,798	-	63,731	363,972	66,103	156,485	13,601	127,784
¹⁴ C activity in metal-bearing wastes (GBq)	1,545	396	1,149	-	-	814	291	523	-	-	114	86	28	-	-	890	395	496	-	-
¹⁴ C activity associated only with metal (GBq)	630	279	350	-	-	526	271	255	-	-	99	81	18	-	-	511	279	232	-	-
Component for HFC (m ³)	37,267	6,664	16,173	1,437	12,993	35,311	6,485	15,168	1,386	12,272	19,504	4,051	9,080	-	6,373	36,397	6,610	15,648	1,360	12,778
Component for non-HFC (m ³)	335,405	59,974	145,559	12,933	116,939	317,803	58,365	136,513	12,478	110,447	175,539	36,463	81,718	-	57,358	327,574	59,493	140,836	12,241	115,005
HFC VRF (Upper)	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
HFC VRF (Lower)	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10
Metal volume (Upper m ³)	9,317	1,666	4,043	359	3,248	8,828	1,621	3,792	347	3,068	4,876	1,013	2,270	-	1,593	9,099	1,653	3,912	340	3,195
Metal volume (Lower m ³)	3,727	666	1,617	144	1,299	3,531	649	1,517	139	1,227	1,950	405	908	-	637	3,640	661	1,565	136	1,278
Total volume after HFC (Upper m ³)	344,721	60,641	147,176	13,076	118,239	326,631	59,014	138,030	12,616	111,674	180,415	36,868	82,626	-	57,995	336,674	60,154	142,401	12,377	116,283
Total volume after HFC (Lower m ³)	339,131	60,641	147,176	13,076	118,239	321,334	59,014	138,030	12,616	111,674	177,490	36,868	82,626	-	57,995	331,214	60,154	142,401	12,377	116,283
Component for Melting (m ³)	372,672	66,638	161,732	14,370	129,933	353,115	64,850	151,682	13,864	122,719	195,043	40,515	90,798	-	63,731	363,972	66,103	156,485	13,601	127,784
Melting VRF (Upper)	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
Melting VRF (Lower)	20	20	20	20	20	20	20	20	20	20	20	20	20	20	20	20	20	20	20	20
Metal volume (Upper m ³)	74,534	13,328	32,346	2,874	25,987	70,623	12,970	30,336	2,773	24,544	39,009	8,103	18,160	-	12,746	72,794	13,221	31,297	2,720	25,557
Metal volume (Lower m ³)	18,634	3,332	8,087	718	6,497	17,656	3,243	7,584	693	6,136	9,752	2,026	4,540	-	3,187	18,199	3,305	7,824	680	6,389
Total volume after Melt (Upper m ³)	74,534	13,328	32,346	2,874	25,987	70,623	12,970	30,336	2,773	24,544	39,009	8,103	18,160	-	12,746	72,794	13,221	31,297	2,720	25,557
Total volume after Melt (Lower m ³)	18,634	3,332	8,087	718	6,497	17,656	3,243	7,584	693	6,136	9,752	2,026	4,540	-	3,187	18,199	3,305	7,824	680	6,389

Key: AC ¹⁴C present as activated material
 CC ¹⁴C present as contamination
 PND ¹⁴C present but not determined
 NP ¹⁴C not present

With a similar format to Table 19, Table 20 presents the volumes of cellulose in future forecast UK LLW following the application of HFC and incineration, both shown separately, for each of the four scenarios described above.

Some particular points of interest are as follows:

- The theoretical maximum reduction in the total cellulose volume for disposal at LLWR following incineration at the lower VRF where all cellulose is suitable for incineration is 100-fold, giving a final disposal volume of only 849 m³. Of this, approximately 626 m³ would be associated with ¹⁴C-bearing wastes having a total activity of 58 GBq.
- The scenario where cellulose can be segregated with a volume of greater than 100 m³ yields approximately 98% of the cellulose in the future forecast LLW inventory. Selecting only those waste streams with a total volume of greater than 100,000 m³ and a cellulose content of greater than 1% yields only around 48% of future forecast cellulose.

The work undertaken to assess the gas migration pathway (McGarry, 2003) showed that the mechanism for the transfer of ¹⁴C through CH₄ gas largely relies upon ¹⁴C being in organic form, as is predominantly the case with contaminated cellulosic wastes. A benefit of incineration, in addition to significant volume reduction for disposal, is that organic species may be converted to inorganic species, thereby reducing the potential for gas transfer of ¹⁴C.

It is of interest to note that around one quarter of future forecast cellulosic arisings do not have associated ¹⁴C activity. The incineration of these wastes is still of benefit, however, as there will be a reduction in post-disposal degradation, hence reduced risk of radionuclide release from the surrounding wastes.

Table 20 Estimated volume changes for future forecast UK LLW cellulose when applying HFC or incineration

	All Cellulosics					All Streams where Cellulosics >20%					All Streams where Stream Volume > 10,000m ³ and Cellulosics > 1%					All Streams where Cellulosics Volume > 100m ³					
	LLWR	AC	CC	PND	NP	LLWR	AC	CC	PND	NP	LLWR	AC	CC	PND	NP	LLWR	AC	CC	PND	NP	
Cellulosics volume (m ³)	84,944	2,927	59,549	217	22,251	62,634	476	49,400	39	12,718	41,089	1,306	33,706	-	6,076	81,304	2,682	56,924	157	21,540	
¹⁴ C activity in cellulose-bearing wastes (GBq)	799	231	568	-	-	45	4	41	-	-	114	86	27	-	-	676	218	457	-	-	
¹⁴ C activity associated only with cellulose (GBq)	58	14	44	-	-	17	1	16	-	-	6	3	3	-	-	43	13	29	-	-	
Component for HFC (m ³)	67,955	2,342	47,639	174	17,801	50,107	381	39,520	31	10,175	32,871	1,045	26,965	-	4,861	65,043	2,146	45,539	126	17,232	
Component for Non-HFC (m ³)	16,989	585	11,910	43	4,450	12,527	95	9,880	8	2,544	8,218	261	6,741	-	1,215	16,261	536	11,385	31	4,308	
HFC VRF (Upper)	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
HFC VRF (Lower)	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10
Cellulosics volume (Upper m ³)	16,989	585	11,910	43	4,450	12,527	95	9,880	8	2,544	8,218	261	6,741	-	1,215	16,261	536	11,385	31	4,308	
Cellulosics volume (Lower m ³)	6,796	234	4,764	17	1,780	5,011	38	3,952	3	1,017	3,287	105	2,696	-	486	6,504	215	4,554	13	1,723	
Total volume after HFC (Upper m ³)	33,978	820	16,674	61	6,230	25,053	133	13,832	11	3,561	16,436	366	9,438	-	1,701	32,521	751	15,939	44	6,031	
Total volume after HFC (Lower m ³)	23,784	820	16,674	61	6,230	17,537	133	13,832	11	3,561	11,505	366	9,438	-	1,701	22,765	751	15,939	44	6,031	
Component for Incineration (m ³)	84,944	2,927	59,549	217	22,251	62,634	476	49,400	39	12,718	41,089	1,306	33,706	-	6,076	81,304	2,682	56,924	157	21,540	
Incineration VRF (Upper)	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7
Incineration VRF (Lower)	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
Cellulosics volume (Upper m ³)	12,135	418	8,507	31	3,179	8,948	68	7,057	6	1,817	5,870	187	4,815	-	868	11,615	383	8,132	22	3,077	
Cellulosics volume (Lower m ³)	849	29	595	2	223	626	5	494	0	127	411	13	337	-	61	813	27	569	2	215	
Volume after incineration (Upper m ³)	12,135	418	8,507	31	3,179	8,948	68	7,057	6	1,817	5,870	187	4,815	-	868	11,615	383	8,132	22	3,077	
Vol after incineration (Lower m ³)	849	29	595	2	223	626	5	494	0	127	411	13	337	-	61	813	27	569	2	215	

Key: AC ¹⁴C present as activated material
 CC ¹⁴C present as contamination
 PND ¹⁴C present but not determined
 NP ¹⁴C not present

6. Conclusions

An assessment of the ^{14}C content of future forecast UK LLW has been undertaken. It was found that around 75% of future forecast ^{14}C activity in the 2007 National Inventory is contributed by only around 18 waste streams, of which the top two contribute 25% and 20% respectively. The large majority of future ^{14}C is forecast to arise after 2060, although annual arising activities prior to this date are significant, with the ^{14}C activity arisings in some years greater than the current LLWR annual limits for ^{14}C . The bulk of ^{14}C -bearing wastes arising after 2060 are graphite from reactor decommissioning and are not expected to be suitable for disposal the LLWR.

The total future forecast ^{14}C activity for LLWR disposal was found to be 1.7 TBq; a four-fold reduction from that calculated using the 2004 National Inventory.

Enquiries made of the 2007 National Inventory data revealed a number of errors for Sellafield LLW '2X' stream fingerprints, reducing the ^{14}C activity for these streams by up to three orders of magnitude. New data were obtained for these waste streams and uploaded to the assessment database for subsequent analysis.

Investigation of the material content of ^{14}C -bearing waste streams destined for LLWR has shown that around 45% of future forecast volume for ^{14}C -bearing wastes is metals, whilst around 13% is cellulose. These material types are of particular interest in the study of ^{14}C migration in the LLWR near-field, since cellulose degradation may lead to ^{14}C -bearing gas generation, whilst corrosion of ^{14}C -activated steel may release activity to groundwater. An assessment was therefore carried out to determine the extent of ^{14}C activity within these and other material types in the LLWR wastes. It was found that the ^{14}C activity associated with cellulose and metals, whilst pervasive through the waste, is predominantly at a low level, in the region of $1.0\text{E}-08$ to $1.0\text{E}-07$ TBq/m³.

Theoretical maximum activity levels in specific material types were calculated by assuming, for each material type within a waste stream, that all the waste stream ^{14}C activity is in that material. It was found that, typically, large increases (i.e. several orders of magnitude) in specific activity from the best estimate across the whole of the waste were only observed for materials with very small volumes, such as complexants; whilst changes in specific activity for materials with large volumes, such as metals, were not significant.

Following identification of a limited number of waste streams contributing the large majority of future forecast ^{14}C , additional data were sought to check the accuracy of the 2007 National Inventory entries. For waste streams currently consigned to the LLWR, WSCDs were obtained, whilst future waste streams were checked through direct contact with the waste producers. The checks consistently found that 2007 National Inventory activities are higher than those stated in the WSCDs; sometimes by several orders of magnitude. Discussion of 2007 National Inventory data with consignors led to a number of potential reductions in the ^{14}C activities due to conservatism in the 2007 National Inventory data being highlighted. Of particular note is the top contributor 7D39/C, for which a process is being developed (ModulOx™) to remove ^{14}C from the waste. If the process is successful the waste will contain minimal amounts of ^{14}C . If the process is found to be unsuccessful the waste will be deemed unsuitable for LLWR disposal.

Magnox decommissioning streams, a source of ^{14}C contamination, are to be reviewed, with potentially large proportions of the volumes being reclassified as VLLW.

It can therefore be concluded that the future forecast total activity of 7.35 TBq for ^{14}C calculated for the lifetime project may be substantially reduced; firstly through a five-fold reduction in estimates from the 2004 to the 2007 National Inventory, and secondly through further investigations revealing a number of errors and conservatism in the 2007 National Inventory data, which may account for about 100 to 200 GBq of ^{14}C activity. Further information may be available from waste producers in 2009 that may

enable a reduced quantitative estimate to be developed. The initial investigations described in this report suggest a potential reduction in the total LLWR future forecast inventory of ^{14}C to less than 800 GBq. However, it is also recognised that uncertainties in the waste streams contributing less than 1% of the ^{14}C inventory, which have not been investigated, may lead to an increase of up to 2 TBq, if the upper band limits as given in the 2007 National Inventory apply.

An assessment of alternative treatment technologies for the processing of waste for LLWR disposal was undertaken to explore the potential impact on the ^{14}C inventory. Incineration of cellulose and melting of metallic wastes were examined against HFC for both waste types. Both were shown to be potentially beneficial, both in reducing future volumes for disposal and in converting ^{14}C to less mobile, inorganic forms, thereby reducing the risk of migration to the environment.

The work undertaken for the Requirement 2 submission used a future vaults ^{14}C inventory of 7.35 TBq, coupled with a cellulosic volume fraction of $3.75\text{E}-02$ to derive an initial evolution rate for ^{14}C -labelled gas of $6.37\text{E}+07 \text{ Bq y}^{-1}$. It has been shown that the majority of ^{14}C is not associated with cellulose; at most having an activity of 800 GBq, but more likely to be in the order of tens of GBq. This represents a direct reduction in the calculated amounts of ^{14}C -labelled gas from cellulose of between 1 and 2 orders of magnitude.

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