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The carbon-14 IPT: an integrated approach to geological disposal of UK wastes containing carbon-14

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ABSTRACT

Carbon-14 is a key radionuclide in the assessment of the safety of a geological disposal facility because of the calculated assessment of the radiological consequences of gaseous carbon-14-bearing species. Radioactive Waste Management Limited has established an Integrated Project Team (IPT) in which partners are working together to develop an holistic approach to carbon-14 management in the disposal system. We have used an 'AND' approach to structure and prioritize our technical work. For a waste stream to be of concern, there has to be a significant inventory, AND carbon-14-bearing gas has to be generated, AND this gas has to be entrained by bulk gas, AND it has to migrate through the engineered barriers, AND it has to migrate through the overlying geological environment (either as gas or in solution), AND there have to be consequences in the biosphere. We are also using this approach to consider alternative treatment, packaging and design options.

KEYWORDS: carbon-14, geological disposal, radioactive waste, Integrated Project Team, IPT.

Introduction

CARBON-14 is a key radionuclide in the assessment of the safety of a geological disposal facility (GDF) for radioactive waste. In particular, the radiological consequences of gaseous carbon-14-bearing species is a potential issue and has been recognized as such in Nirex report N/122 (Nirex, 2005), and in the generic Disposal System Safety Case (DSSC) (Nuclear Decommissioning Authority, 2010*a*).

Radioactive Waste Management Limited (RWM) has been carrying out a range of research and assessment tasks on carbon-14 to improve the understanding of the expected evolution of the disposal system and the consequences for the calculated annual risk. Much of this work is summarized in the gas status report (Nuclear Decommissioning Authority, 2010*b*), the radionuclide behaviour status report (Nuclear Decommissioning Authority, 2010*c*), and in the R&D Programme Overview document (Nuclear Decommissioning Authority, 2011). However, following publication of the generic DSSC, it was decided to adopt a collaborative approach to tackling issues related to carbon-14 by establishing an Integrated Project Team (IPT), in which the partners work together to develop an holistic approach to carbon-14 management in a geological disposal facility.

The key inputs of improved understanding through models and data contribute to an updated assessment and consideration, if required, of alternative treatment, packaging or design options. These may in turn lead to requirements for

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further improvements in understanding, models and data.

The Phase 1 report was issued at the end of 2012 (Nuclear Decommissioning Authority, 2012). We are now undertaking the work in Phase 2 of the project, which will be reported in 2016. The focus of this work programme is to develop a better understanding of: (1) the inventory of the important streams; (2) the source terms for gaseous release from graphite, steels and packages containing reactive metals; (3) whether the gas will migrate through the generic geological environments being considered; and (4) the consequences in the biosphere of any gases released.

The results of some of this work are discussed in the remainder of this paper.

Key generation and migration processes: The 'AND' approach

The key processes that could result in a significant radiological consequence from carbon-14 are shown in Fig. 1. Gas can be generated in the near field of a GDF if water is able to interact with the wastes. The main gas generation processes are corrosion of metals (either metals in the wastes or metals in the containers), degradation of organic materials and radiolysis. The bulk of the gas produced will be hydrogen, and it may act as a carrier for other gases if it is generated in sufficient quantity. Other gases that may be generated include methane and carbon dioxide, and these gases could contain carbon-14. Carbon dioxide is likely to be retained within the engineered barrier system through carbonation of cementitious materials in the GDF (for example, present in encapsulation grouts and backfill). However, methane is likely to be either dissolved or entrained within a separate bulk gas phase. Whether gas migrates to the biosphere is determined by the features of the geological environment, in both the host rock and the overlying sequences. If gas is able to reach the biosphere, it could be released directly as methane or converted to carbon dioxide by microbes in the soil zone. This carbon dioxide would then be taken up by plants and enter the food chain, leading to a consequent dose to any exposed groups or potentially exposed groups. Further information on the important processes can be found in Nuclear Decommissioning Authority (2010*b*) and Rodwell *et al.* (1999).

Systematic approaches to demonstrating safety in the nuclear industry have been developed over several decades; they include the structured approach to hazard identification known as HAZOP (hazard and operability) in nuclear safety (Tyler *et al.,* 2008) and the FEP (Features, Events and Processes) approach that is used in radioactive waste disposal assessments (Bailey and Billington, 1998). The FEP approach has informed the UK's understanding of the potential contributors to the evolution and migration of carbon-14 in disposal concepts which are considered viable in the UK's geology (Nuclear Decommissioning Authority, 2010*a*). However, a novel approach was developed herein, recognizing that successive specific features, events and processes would be required in order for a potentially detrimental uptake to occur in the surface environment. Our integrated 'AND' approach, illustrated in Fig. 2, integrates our understanding of the waste inventory and its chemical form, its evolution with time, migration of carbon-14 through the engineered

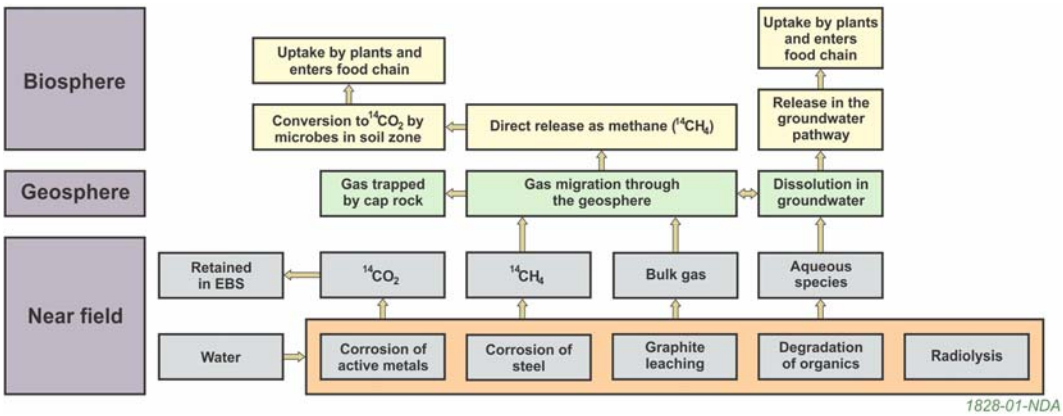

FIG. 1. The key processes for the generation and migration of ^{14}C-bearing gases. Figure published with permission of the NDA.

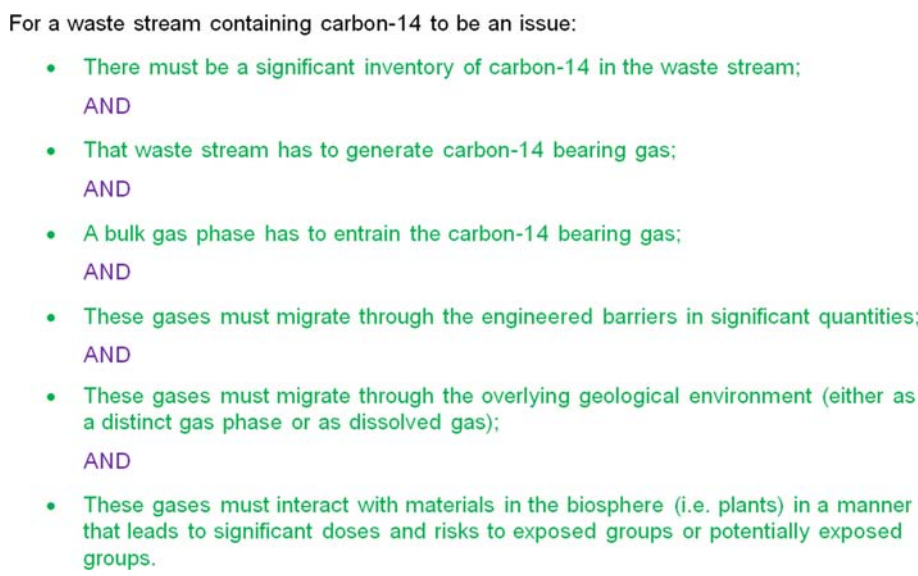

FIG. 2. The integrated technical approach (the AND approach).

barriers and the geosphere and uptake in the environment, recognizing the combination of events required for any significant radiological uptake to occur in the future.

The assessed risk from the migration of carbon-14 in groundwater is generally found to be below the risk guidance level, because the ground water travel time is long in comparison with the half-life of carbon-14 (Nirex, 2005). However, because there is interaction between carbon-14 in gas and groundwater, it may be appropriate to consider them together within a total system model[1].

The IPT has used this approach as a way of considering the problem comprehensively and to underpin the prioritization of the technical work. We have used the 'AND' approach for each group of waste streams in order to break the problem down in a manageable way. We have also used it to underpin our integration activities, in particular the approach to alternative treatment packaging and design options.

Phase 1 results

Ignoring any potential benefits from the geosphere, the calculated consequences of carbon–14 in the Phase 1 work are dominated by the corrosion of irradiated reactive metals (in the operational[2] and early post-closure period), and the corrosion of irradiated stainless steel and leaching of irradiated graphite (in the longer post-closure period). The generation rates for unshielded ILW are shown in Fig. 3 (Nuclear Decommissioning Authority, 2012).

Inventory

The Phase 1 work (Nuclear Decommissioning Authority, 2012) used the 2010 Estimated Derived Baseline Inventory; there are ~9880 TBq (~10^{16} Bq) of carbon-14 in that inventory. It comes from activation of nitrogen-14, carbon-13 and oxygen-17 'precursor' species. The half-life of carbon-14 is 5730 years, so if it is retained for sufficient time, it will decay. If carbon-14 is released as gas and migrates directly to the biosphere then there could be an issue, as the time for gas to migrate may well be significantly less than the half-life.

The vast majority of the carbon-14 originates from nuclear reactors and the reprocessing of spent fuel and other fuel element components. Table 1 shows the major contributors to the total carbon-14 inventory. The wastes have been divided into material categories – graphite, steels, reactive

[1]A total system model is a simplified model used to calculate overall performance measures of the geological disposal system, such as radiological dose and risk.

[2]In the Phase 1 work, the operational period is up to around 2150.

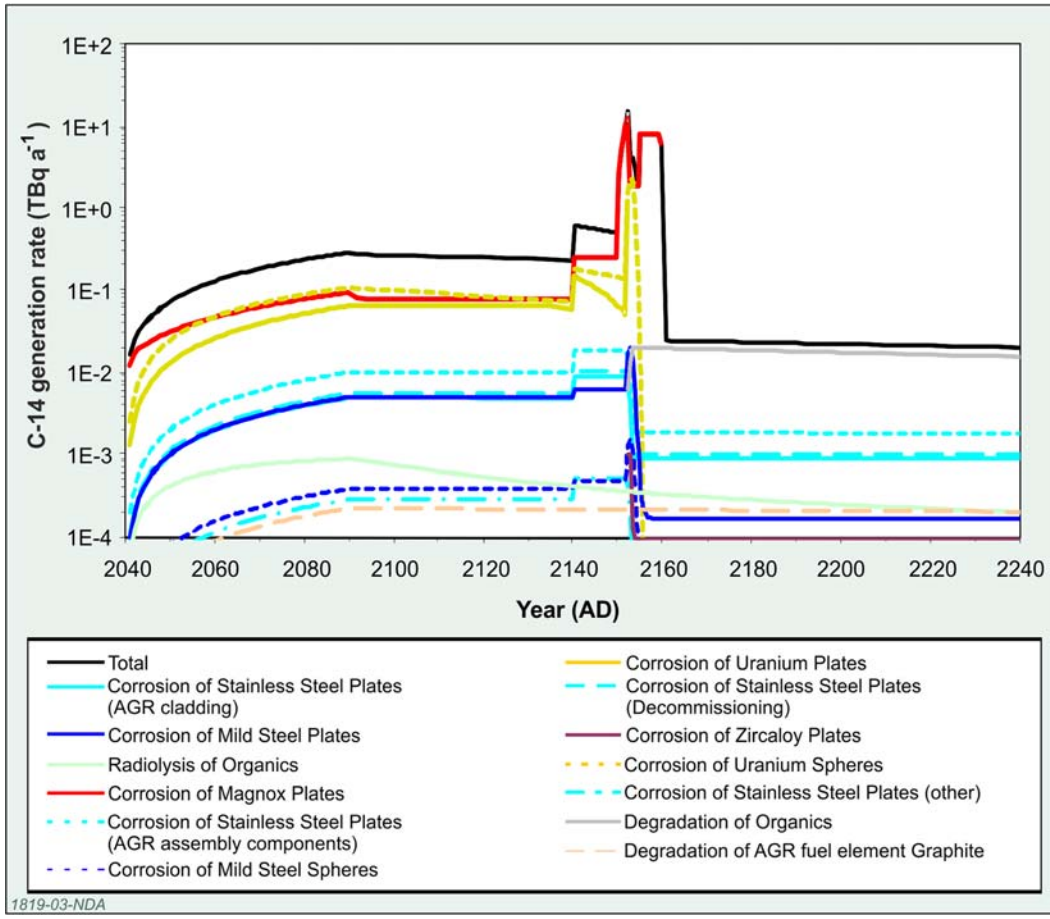

FIG. 3. The Phase 1 results for unshielded ILW. Figure published with permission of the NDA.

metals (Magnox plus uranium) and other wastes – and within these categories into separate waste groups. The categories and waste stream groups have been informed by discussions within the IPT.

The focus of the work of the IPT has been on the main intermediate-level waste streams (ILW) in Table 1, namely irradiated graphite, irradiated steels and irradiated reactive metals. Of the other wastes, the main contributors to the inventory are certain ILW organic wastes from GE Healthcare, which are expected to be incinerated, and spent fuel, which is expected to be disposed in containers that retain the wastes for timescales significantly longer than the half-life of carbon-14.

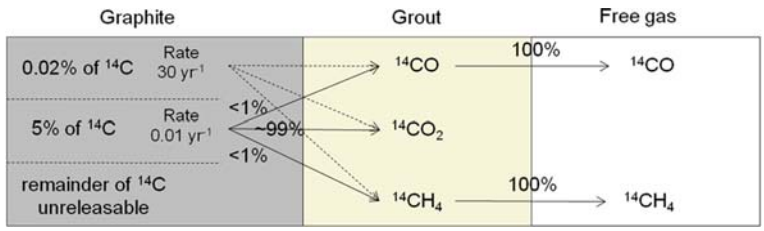

FIG. 4. The revised model for the release of carbon-14 from graphite. Figure published with permission of the NDA.

TABLE 1. Major components of carbon-14 in the 2010 Estimated Derived Inventory (EDI). Baseline inventory showing enhancements.

Category – Waste stream group	Total C-14 activity (TBq)	Contribution to total C-14 activity (%) [4]
Graphite - ILW core graphite	7,132	72.2
Graphite - ILW AGR fuel element graphite	45	0.5
Graphite - ILW Magnox fuel element graphite	6	0.1
Steels - ILW AGR stainless steel fuel cladding	28 (+143) [1]	1.7
Steels - ILW AGR stainless steel fuel assembly components	356	3.6
Steels - ILW stainless steels from reactor decommissioning	226	2.3
Steels - ILW other ferrous metal decommissioning wastes	197	2.0
Steels - ILW other ferrous metal reactor wastes	28	0.3
Reactive metals - ILW reactive metals (Magnox and uranium)	66 (+50)	1.2
Other wastes - ILW organics from GE Healthcare [2]	560	5.7
Other wastes - Other ILW	67	0.7
Other wastes - Spent fuels	926	9.4
Other wastes - LLW core graphite	6	<0.1
Other wastes - Miscellaneous not assigned [3]	40	0.4
Total C-14 in 2010 EDI Baseline Inventory	9683 (+193)	100

(1) Bracketed figures show the impact of enhancement of the 2010 EDI. These enhancements have been carried forward from the 2007 Derived Inventory.
(2) ILW organics from GE Healthcare may not be disposed to the GDF.
(3) This group comprises those waste streams that contain low concentrations of carbon-14 but do not fit into any of the designated waste stream groups.
(4) Includes enhancements.

The IPT is improving our understanding of the main components of the inventory of carbon-14. This improved understanding will be reflected in the 2013 Derived Inventory, which will form the basis of the Phase 2 calculations.

Irradiated graphite

A number of experimental studies have been undertaken on the release of carbon-14 from irradiated graphite. Although graphite is a very stable material, small quantities of carbon-14 have been measured to be released from graphite in contact with water. In the most recent experiments irradiated graphite from the Oldbury Magnox station has been used (Baston *et al.,* 2012, 2014). This work concluded that under baseline conditions (anoxic, under pH 13 solution, ambient temperature – conditions that are typical of groundwaters in a facility for the disposal of ILW), the predominant carbon-14 release was to the solution phase. About 1% of the released carbon-14 was present in the gas phase, with the majority of that in methane, along with small quantities of carbon monoxide.

We have compared the results of our experiments with other work, and we conclude: (1) a fraction of the C-14 was released rapidly; (2) a proportion was released more slowly; (3) some was not released at all; (4) most was released into the aqueous phase; and (5) a small amount was released as gas.

On the basis of this we have developed a revised model of the release of carbon-14 from graphite, which is represented in Fig. 4 (Baston *et al.,* 2014).

The results of the revised model are compared with the results of the earlier model in Fig. 5. It is seen that the maximum release rate is unchanged, whereas the duration of the release is much reduced. This arises because a proportion of the carbon-14 remains in the graphite. The figure also shows a number of variants looking at the sensitivity to some of the key parameters.

We are currently considering the option of disposing of core graphite in separate vaults. Certain options offer the possibility that a gas phase may not be associated with graphite wastes if the quantities of bulk gas are reduced, for example if the graphite wastes are segregated and appropriate packaging used.

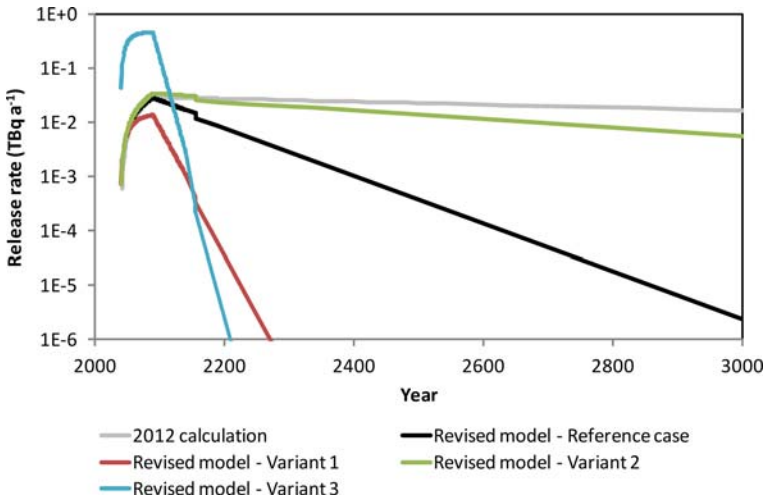

FIG. 5. Results from the revised model for the release of carbon-14 from graphite. Figure published with permission of the NDA.

Irradiated steels

Analysis of the carbon-14 inventory in current irradiated stainless steels wastes, has identified two streams, namely 2F03/C (Encapsulated AGR cladding) and 2F08 (AGR stainless steel fuel assembly components) as being the major contributors to the current inventory. In addition, certain future decommissioning waste streams from possible new-build reactors have also been identified as having a significant inventory (the identification of these streams came after the preparation of Table 1). Our work is focused on obtaining better data for these streams.

We are also participating in the EU CAST (CArbon-14 Source Term) project. As part of this project, we are reviewing the international understanding of the very low corrosion rates of stainless steel. We are also going to undertake some long-term experiments on the release of carbon-14 from irradiated steels as part of CAST, using a similar experimental approach to that used in the successful graphite experiments. The bulk of the gas generated is expected to be hydrogen, but this could act as a carrier gas for small quantities of, for example, carbon-14 bearing methane.

Irradiated reactive metals

The key waste streams containing irradiated reactive metals come from Sellafield; a modest number of streams contribute most of the carbon-14 inventory. In the Phase 1 results, these wastes gave the highest release rates in the operation and early post-closure periods.

In the current work being undertaken for RWM: (1) the release of carbon-14 from irradiated Magnox is being measured; (2) package-scale modelling is being undertaken to understand the environment in which the reactive metal corrodes; and (3) further data are being sought on the effects of chloride on the Magnox corrosion rate.

From the work, we have observed that: (1) in most cases, there is sufficient water in the package itself to allow a significant proportion of the Magnox to corrode; and (2) the corrosion rate and therefore the time taken to corrode depend on the temperature and availability of chloride[3].

A simple model based on the corrosion rates and typical waste dimensions provides indicative corrosion times given in Table 2.

From this we see it is important to understand whether the corrosion rates being used are pessimistically high, and whether significant quantities of chloride enter the package. We are also considering possible alternative approaches, and developing a total system model to facilitate a structured consideration of uncertainty.

[3]Although chloride is not expected to be present in the waste or grout it may enter the packages after closure of the GDF and resaturation of the facility, because of its presence in many deep groundwaters.

TABLE 2. Indicative corrosion times for Magnox for different chemical conditions.

Environmental conditions	Corrosion time
GDF conditions, chloride present	~10 years
35°C, No chloride	~750 years
25°C, No chloride	~2900 years
15°C, No chloride	~11,000 years

Formation and migration

In this area our focus is currently on:

(1) Understanding under what circumstances would a gas phase form. This depends on the geological environment and, in particular, how much groundwater is available to dissolve the gas. It also depends on how much gas is generated from particular wastes, which in turn depends on the nature of the wastes and the quantity of metal in their packaging.

(2) Compiling an evidence base on carbonation in the near field and on microbial interactions in the near field. In particular $^{14}CO_2$ is expected to react with cementitious materials in both the waste grout and vault backfill (carbonation).

(3) Understanding the key geological features in the generic environments that determine the residence time for gas in the geosphere before it reaches the biosphere, and the areas over which it is expected to be released if it does reach the biosphere. Ultimately, it is recognized that gas is a site-specific issue. The residence time and release area depend on the properties of the host rock (permeability and porosity, and in particular the two-phase flow properties), and also on the properties of the overlying sequences and whether there are sequences that might act as a cap preventing, or delaying, the release of gas. An understanding of the key features and properties enables the migration of gas to be modelled (Hoch and James, 2012).

These are being considered in the context of the three generic geological environments being considered by RWM (higher strength rock, lower strength sedimentary rock and evaporites). Lower strength sedimentary rocks and evaporites would

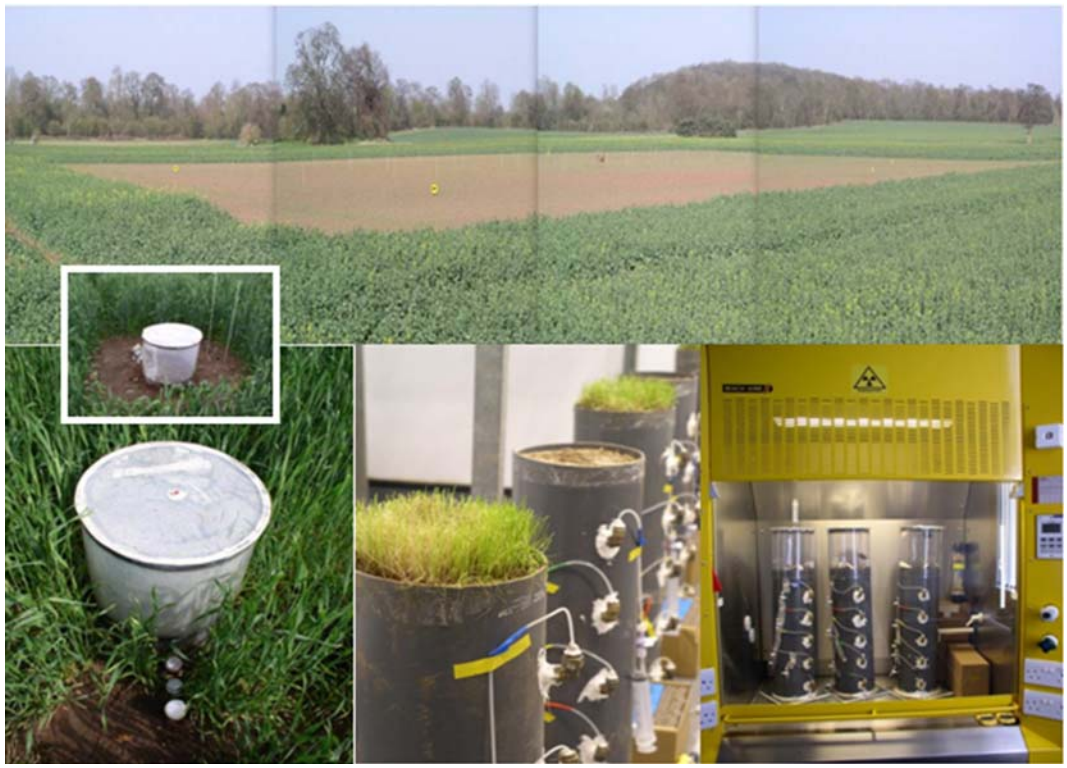

FIG. 6. Field and laboratory experiments examining the oxidation of methane.

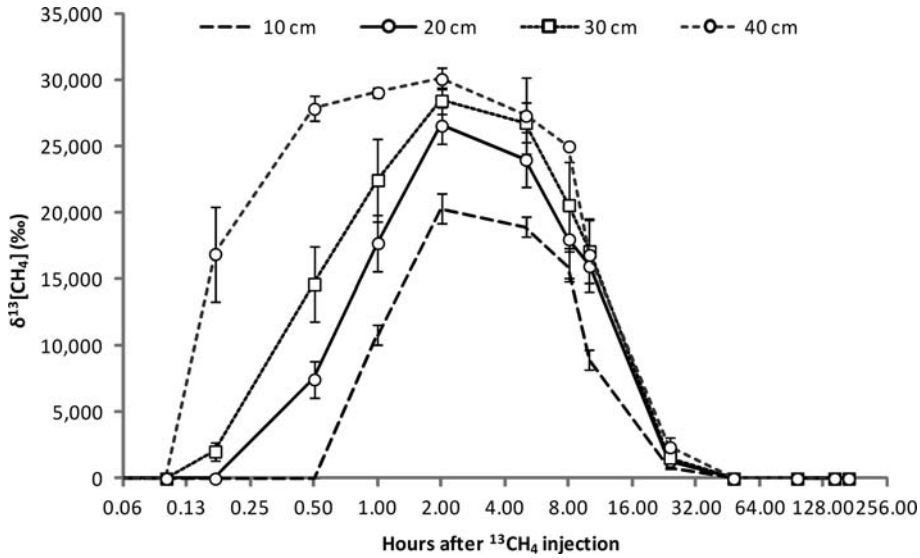

FIG. 7. Laboratory results. Average $\delta^{13}CH_4$ measured within gas samples obtained from four depths in LE 4. Figure published with the permission of the NDA.

TABLE 3. A comparison of the flux to dose conversion factor for gaseous $^{14}CH_4$ from the revised assessment model with the earlier value.

Assessment	GPA(03) update	New model
Flux to dose conversion factor (Sv yr^{-1}) per (Bq m^{-2} s^{-1})	0.6280	0.0225

offer a significant barrier to gas migration. They might also limit water flow into the GDF, thereby limiting the rate of metal corrosion.

Biosphere

A series of laboratory and field experiments have been undertaken by the University of Nottingham to investigate the transport and retention of $^{13}CH_4$ (as a stable analogue for the radioactive $^{14}CH_4$) in agricultural soil. Radioactive $^{14}CH_4$ was also used in one of the laboratory experiments.

The focus of these experiments is to understand the extent that methane, released to the bottom of the vadose zone, is oxidized to form carbon dioxide. Some experiments studied the soil processes with a growing crop, whereas others had no crop. To examine two different crops, the field experiments used ryegrass in the first year (2011) and spring wheat in the second (2012). Antecedent measurements were taken before methane injection, including various soil properties and soil methane concentrations at a number of depths. Methane fluxes into or out of the soil were estimated using a fixed volume head space chamber in which the air was continuously stirred. Then methane labelled with ^{13}C was injected at a depth of about 50 cm, and samples were taken throughout the soil profile a number of times following injection. Head space measurements were again taken at a number of times after injection to estimate fluxes of methane and carbon dioxide. The experiments are reported in Atkinson *et al.* (2011, 2013, 2014) and Shaw *et al.* (2013). The set up of these experiments is illustrated in Fig. 6. Some typical results are shown in Fig. 7. The results of these experiments have been modelled using both simple and detailed models (Hoch and Shaw, 2014).

In the light of the results of these experiments, the model that is used to assess the consequences of a release of $^{14}CH_4$ into the subsoil has been reviewed, and a revised assessment model developed. The main conclusion from the work is that most of the radioactive methane migrating from a deep repository is likely to be converted to radioactive carbon dioxide in the soil.

The experiments and the associated modelling have shown that the original cautious assumption of complete conversion of methane oxidation to carbon dioxide should not be changed. The approach is consistent with that adopted by the Low Level Waste Repository (LLWR) (Sumerling, 2013). Improvements in the assessment model arising from a more detailed treatment of canopy and above-canopy processes (see Hoch, 2014) have resulted in a reduction by a factor of just under 30 in the model for converting a flux of methane to a biosphere dose compared to the one used in the 'GPA(03) Update' (Hoch *et al.,* 2008). The values are given in Table 3.

A revised methodology has also been developed for use in the Operational Environmental Safety Assessment. This updates the original methodology, which was intentionally cautious.

Conclusions

The Phase 2 work of the IPT is currently underway. Good progress is being made in terms of:

(1) Examining the justification of the underpinning information. An example of this is the inventory of carbon-14 to be disposed in the GDF; for some parts of the inventory, the data will be improved, for others better justification will be provided.

(2) Providing an improved understanding of how the system will evolve. An example of this is the package-scale modelling that is being undertaken for reactive metals.

(3) Providing improved models of important parts of the system, for example the model of the graphite source term.

(4) Examining whether there are practicable alternative options for the treatment, packaging, design or disposal of the wastes.

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