



An approach to modelling the impact of ^{14}C release from reactor graphite in a geological disposal facility

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ABSTRACT

Carbon-14 (C-14) is a key radionuclide in the assessment of a geological disposal facility (GDF) for radioactive waste. In the UK a significant proportion of the national C-14 inventory is associated with reactor-core graphite generated by the decommissioning of the UK's Magnox and AGR reactors.

There are a number of uncertainties associated with the fate and transport of C-14 in a post-closure disposal environment that need to be considered when calculating the radiological impacts of C-14-containing wastes. Some of these uncertainties are associated with the distribution of C-14-containing gaseous species such as $^{14}\text{CH}_4$ and $^{14}\text{CO}_2$ between the groundwater and gaseous release pathways. As part of the C14-BIG programme, a modelling framework has been developed to investigate these uncertainties. This framework consists of a biogeochemical near-field evolution model, incorporating a graphite carbon-14 release model, which interfaces with a geosphere/biosphere model. The model highlights the potential impact of the microbial reduction of $^{14}\text{CO}_2$ to $^{14}\text{CH}_4$, through the oxidation of H_2 , on C-14 transport. The modelling results could be used to inform the possible segregation of reactor graphite from other gas-generating wastes.

KEYWORDS: carbon-14, graphite, gas generation, geosphere, dose assessment.

Introduction

APPROXIMATELY 80,000 tonnes of irradiated graphite will arise from the decommissioning of Magnox and AGR reactors (NDA, 2011, 2014). This graphite contains the majority of the UK intermediate-level radioactive waste C-14 inventory (NDA, 2012). C-14 is a key radionuclide in the assessment of a geological disposal facility (GDF) for UK radioactive waste and the radiological consequences of gaseous C-14 transport in particular have been recognized as a potential issue (NDA, 2012).

There are uncertainties associated with the long-term release of C-14 from irradiated graphite which are related to the release mechanisms, the chemical forms, and the total releasable fraction of the C-14 inventory. In particular the partitioning of C-14 between aqueous and gaseous phases could have a significant impact on the transport of C-14 bearing species.

The release of C-14 via a gas release pathway in a fractured higher-strength host rock is highly dependent on the volumes of non-radioactive gases generated within a GDF, as these gases have the ability to drive the transport of the smaller volumes of C-14 bearing gases. Microbiological and corrosion processes may generate gas within a disposal facility and the associated biogeochemical

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processes have been addressed in detail by several studies (Humphreys *et al.*, 1997; Graham *et al.*, 2003; Suckling *et al.*, 2011). The most mobile C-14 species is likely to be CH₄ as this is not subject to precipitation reactions that will retard the migration of C-14-labelled CO₂. If C-14-labelled CH₄ reaches the biosphere it will be subjected to microbial oxidation to C-14 CO₂; once converted to CO₂ the C-14 is available for root and foliar uptake by plants (Hoch *et al.*, 2014).

The mechanisms associated with the migration of radioactive gases are strongly influenced by the host rock and could be described by following a two-phase flow approach (Kuitunen, 2011; Schwartz, 2012). It is difficult, however, to incorporate a complex two-phase flow modelling into assessment-level models. Therefore, an alternative approach, which is more suitable for screening studies, is to describe the advection and dispersion processes in the geosphere with a simplified mass pathway (Limer *et al.*, 2010; Towler *et al.*, 2012).

The impact of C-14 release from graphite in geological disposal has been addressed in several studies. Bracke and Muller (2008) discussed possible scenarios for the release pathways and the associated processes towards a less conservative approach in the release of C-14 from a low-level waste repository. Smith *et al.* (2012) presented a comparison of five models used to predict uptake of C-14 to agricultural crops, within the international BIOPROTA framework. Towler *et al.* (2012) presented an analysis related to UK graphite in a deep repository during the EC CARBOWASTE project. The analysis presented in those studies demonstrated that, in principle, it should be possible to dispose safely of irradiated graphite wastes in isolation, in a wide range of disposal systems, including near-surface, shallow and deep

geological disposal, and in a wide range of host rocks.

As part of the C14-BIG programme, a modelling framework has been developed that goes beyond that modelled by Towler *et al.* (2012) by integrating a range of gas generation processes with recent insights into the speciation of C-14 leaching from reactor graphite (Baston *et al.*, 2014). This framework consists of a biogeochemical near-field evolution model, incorporating a graphite C-14 release model, which interfaces with a geosphere/biosphere model developed in a GoldSim (2010) simulation environment.

Model description

Near-field processes

C-14 release from reactor graphite

The release of C-14 from graphite is modelled using the approach presented recently by Baston *et al.* (2014). The conceptual model considers three fractions: a rapidly releasable fraction loosely bound to the graphite surface; a slowly releasable fraction from the graphite porosity; and a retained fraction that is immobilized in the graphite structure. The C-14 released is further speciated into CO₂, CO and CH₄ before being released into the aqueous or gaseous phases. The conceptual model is illustrated in Fig. 1 and the release rate is calculated as:

$$q = (k_{ca}F_{ca}e^{-k_{ca}t} + k_{cc}F_{cc}e^{-k_{cc}t})A_0e^{-\lambda_c t} \quad (1)$$

where *q* is the release rate of C-14 from graphite [TBq/y], *k_{ca}* and *k_{cc}* are the rate constants for the rapid and the slow release [y⁻¹], respectively, *F_{ca}* and *F_{cc}* are the rapidly and the slowly releasable fraction, respectively, *A₀* is the initial activity of C-14 in the graphite [TBq], *λ_c* is the radioactive decay rate of C-14 [a⁻¹] and *t* is time [y].

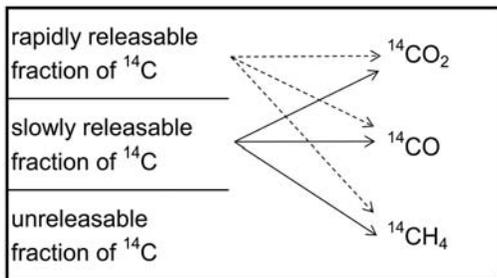


FIG. 1. Conceptual model of C-14 release from graphite (adapted from Baston *et al.*, 2014).

Corrosion

The corrosion of steel is modelled under anaerobic, saturated conditions via the following reaction scheme (Suckling *et al.*, 2011):



Steel corrosion is modelled by zero-order kinetics and is given by:

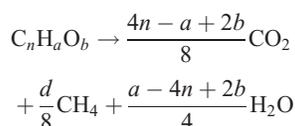
$$-\frac{dQ}{dt} = \frac{\rho r A}{M} \quad (2)$$

where *Q* is the mass of steel [mol], *ρ* is the density

of steel [kg/m^3], M is effective molecular mass of steel [kg/mol], r is the corrosion rate under anaerobic conditions [m/y] and A is the surface area of steel [m^2].

Microbial degradation

Cellulose is the only class of organic waste considered within the model. The microbial degradation of polymeric organics such as cellulose follows a generic sequence of events (Rittmann and McCarty, 2001), where first the polymer is converted into soluble intermediates. These are then subjected to microbial oxidation reactions depending on the prevailing geochemical conditions. The microbial degradation of polymeric substrates and the subsequent generation of gas is a rather complex process involving an array of microbial species. Representation of this process can be simplified by the fact that the conversion of polymeric substrates to their soluble intermediates is often the rate-limiting factor in the gas-generation process (Rittmann and McCarty, 2001), allowing the microbial oxidation of polymers to be coupled directly to the reduction of the relevant terminal electron acceptors. This approach has been applied in other modelling studies (McNab and Narasimham, 1994; Postima and Jacobsen, 1996; Suckling *et al.*, 2011), because it simplifies the model by reducing the number of kinetic parameters required. In the model reported here, organic degradation is modelled by the following reaction:



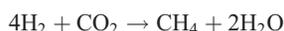
where $d = 4n + a - 2b$ (for cellulose: $n = 6$, $a = 10$, $b = 5$, $d = 24$).

The degradation of cellulose is modelled by first-order reaction kinetics:

$$-\frac{dC}{dt} = kC \quad (3)$$

where C is the mass of polymer [mol] and k is the degradation rate constant under anaerobic conditions [y^{-1}].

Microbial metabolism of corrosion-derived hydrogen follows first-order reaction kinetics and is coupled to the reduction of carbon dioxide:



The microbial consumption of hydrogen and the generation of hydrogen from corrosion have

independent reaction rates. If the rate of microbial hydrogen metabolism is greater than that of corrosion and the associated hydrogen generation, then the rate of corrosion becomes the limiting process.

Microbes utilize part of the degraded organic material to produce new biomass of the composition $\text{C}_5\text{H}_7\text{O}_2\text{N}$. Microbes are also subjected to natural turnover as cells have finite lifetimes and the model assumes that a fraction of the dead biomass is recycled to the organic material. The production rate of biomass is controlled by the degradation rate of the organic material and the decay rate of biomass, and is given by:

$$\frac{dX}{dt} = Y \frac{dC}{dt} - DX \quad (4)$$

where X is the biomass [mol], Y is the biomass yield coefficient, and D is the biomass decay rate [y^{-1}].

The microbial groups required to catalyse the processes above are assumed to be present within the repository. This can be justified on the basis that microbes will enter the repository during the construction and operational phases, and in the waste and the groundwater entering the repository.

Microbial activity in the near field of a cementitious GDF will be influenced by the ambient pH conditions. In an homogenous, highly alkaline environment ($\text{pH} > 12.5$) (NDA, 2010a), microbial activity is unlikely (Humphreys *et al.*, 2010), and in this case only corrosion and cellulose hydrolysis will proceed. In a heterogeneous near field, lower pH niches (between 10 and 12.5) may occur, allowing microbial activity to take place. Microbial activity at pH values < 12.5 has been demonstrated by experimental work carried out as part of the C-14 BIG project (Rout *et al.*, 2015) which has shown that microbially driven organic degradation and associated methane generation can occur at pH 11.0 in completely mixed systems.

Chemical processes

The model does not simulate an evolving near-field chemistry or take into account temperature variations, rather it is a scenario-based approach which is employed to simulate qualitatively different near-field evolutions. In addition, the model does not take into account any potential disequilibria between C-14 and C-12 and the model implicitly assumes initial isotopic equilibrium in all phases. One key process considered by the model is carbonation, where dissolved CO_2 has the potential to be retained

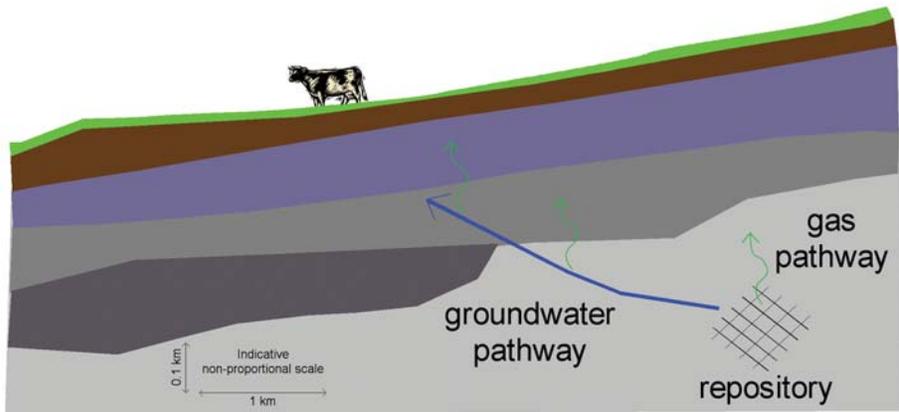


FIG. 2. Groundwater and gas pathways in the geosphere.

in the near field via carbonation reactions with cementitious materials within the GDF (NDA, 2010a). Carbonation is important because it has the potential to retain C-14 in the near-field through the precipitation of C-14-bearing CO_2 .

Additional aspects of near-field modelling

In the modelling approach presented here, the generation of biogenic and C-14-bearing gases in a GDF is influenced by microbial activity, metal corrosion and the release of C-14 from graphite. The migration of C-14 to the geosphere/biosphere environment follows a groundwater pathway or a gas pathway and is influenced by the magnitude of groundwater flow and the partitioning of gas between the solution and the gaseous phase. The relevant modelling assumptions are discussed below.

The model assumes, for simplicity in run time, constant (in time and space) values of rock permeability and hydraulic gradient and uses Darcy's law to estimate groundwater inflow and outflow in the near field. C-14-bearing species dissolved in the groundwater entering the far field from the near field constitutes the groundwater release pathway.

The gas generated in the near field is partitioned between the solution and the gas phase using Henry's law. The gas phase is hosted in an assumed headspace within the repository (Humphreys *et al.*, 1997; Graham *et al.*, 2003). The aqueous phase is in equilibrium with the headspace gases and it is assumed that there is a perfect mixing between the gases both in the headspace and the aqueous phase. The carbonate chemistry associated with carbon dioxide solubility is not modelled explicitly. Rather

the carbonation process is modelled as a sink term for carbon dioxide removal, an approach consistent with other gas generation models (Rodwell, 2004). A fractured, higher-strength host rock is assumed to host the repository and as such is assumed not to represent a significant barrier to gas migration (NDA, 2010c). Gas is released to the far field via the headspace based on the mean near field gas pressure, which is determined by assuming that hydrostatic conditions exist. This release of C-14-bearing gases to the far field constitutes the gaseous release pathway.

Far-field modelling

The migration of C-14 from the near field follows the groundwater or the gaseous release pathway through the geosphere before entering the biosphere environment. A detailed representation of the geosphere/biosphere system is beyond the scope of the model presented here. The model presented here simplifies these processes in order to understand the impact of a number of near-field assumptions on the transformation and release of C-14 with the aim of informing future methodological developments.

The groundwater and gas pathway of C-14 from the repository to the biosphere is shown in Fig. 2. Along the groundwater pathway, the model considers a constant groundwater flow and advection and dispersion processes through a homogeneous and isotropic geological medium. Groundwater abstraction is modelled from a near-surface aquifer system. The model allows dissolved C-14 species to be released into the gas phase as the groundwater

MODELLING IMPACT OF C-14 RELEASE FROM GRAPHITE IN GEOLOGICAL DISPOSAL

TABLE 1. Wastes in the assumed repository and the parameters of the modelling framework.

Parameter	Value	Reference
Graphite radioactivity [TBq]	6000	Limer <i>et al.</i> (2010)
Cellulose mass [Mmol]	9.3	NDA (2010c)
Steel mass [Mmol]	1298	NDA (2010c)
Rapidly releasable fraction of C-14	1.5×10^{-4}	Baston <i>et al.</i> (2014)
Slowly releasable fraction of C-14	5×10^{-2}	Baston <i>et al.</i> (2014)
Fraction of C-14 released as CH ₄	10^{-2}	Baston <i>et al.</i> (2014)
Fraction of C-14 released as CO	5×10^{-3}	Baston <i>et al.</i> (2014)
Rate of rapid release of C-14 [a ⁻¹]	50	Baston <i>et al.</i> (2014)
Rate of slow release of C-14 [a ⁻¹]	2.5×10^{-3}	Baston <i>et al.</i> (2014)
Steel corrosion rate[μm/a]	10^{-2}	
Surface area of steel [km ²]	10	
Cellulose degradation rate [a ⁻¹]	5×10^{-4}	Walke <i>et al.</i> (2011)
Hydrogen metabolism rate [a ⁻¹]	250	Walke <i>et al.</i> (2011)
Biomass yield coefficient [-]	0.23	Walke <i>et al.</i> (2011)
Biomass decay rate [a ⁻¹]	10	Walke <i>et al.</i> (2011)
Recycling fraction of dead biomass	0.9	Walke <i>et al.</i> (2011)
H ₂ solubility [mol/m ³ /atm]	0.79	
CH ₄ solubility [mol/m ³ /atm]	1.42	
CO ₂ solubility [mol/m ³ /atm]	34.17	
Length of repository [m]	1500	NDA (2010b)
Width of repository [m]	1000	NDA (2010b)
Height of repository [m]	20	
Height of headspace [m]	0.04	
Permeability in the near field [m ²]	10^{-16}	
Permeability in the far field [m ²]	3.3×10^{-12}	
Hydraulic gradient of groundwater [-]	0.001	Limer <i>et al.</i> (2010)
Porosity in the near field [-]	0.1	
Porosity in the far field [-]	0.28	
Dispersivity [m]	10	
Diffusivity [m ² /s]	4.3×10^{-10}	
Length of the groundwater pathway [m]	6000	
Release area of the gas pathway [m ²]	18×10^6	
Flow area of the groundwater pathway [m ²]	20,000	
Flow area of the gas pathway [m ²]	9	

approaches the surface because water pressure decreases along with associated solubility of C-14-bearing gases. Gaseous species released from the groundwater are assumed to travel to the biosphere through faults in the higher-strength host rock following mostly a vertical elevation. The migration of the free gas phase generated in the near field and in the groundwater pathway is controlled by a constant gas-flow rate depending on rock permeability, flow area and pressure difference according to the Darcy flow equation.

The biosphere environment is simulated by using three discrete model compartments: the soil-plant, the animal uptake and the human uptake. The soil-plant compartment is based on the enhanced RIMERS model (Thorne, 2005) and is interfaced

both with the gas and the groundwater pathway. Within the soil compartment, ¹⁴CH₄ in the free gas phase is assumed conservatively to be fully metabolized by microbes to ¹⁴CO₂. The animal and the human uptake compartments are based on the ingestion rates of particular food sources and the C-14 dose coefficients; these are only interfaced with the groundwater pathway. The biosphere is modelled in line with the approach described by Limer *et al.* (2010).

Scenarios description

The modelling framework is applied to an assumed repository at a depth of 650 m containing intermediate-level radioactive wastes based on the

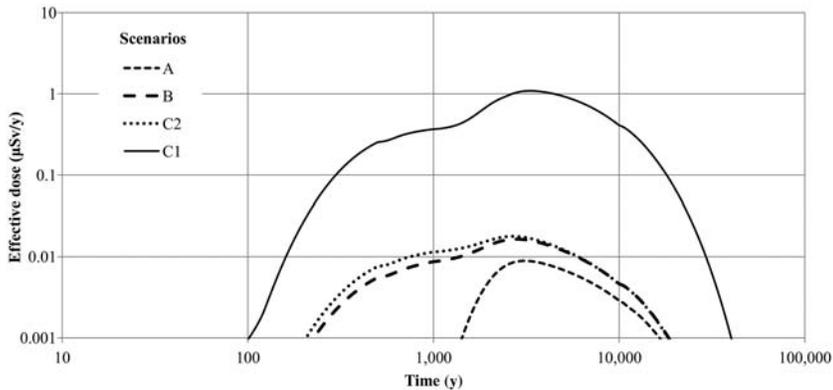


FIG. 3. Radiological effective dose for the scenarios in the near-field environment.

UK inventory. Fully saturated and anaerobic conditions are assumed from the beginning of the simulation as only the post-close environment is considered here. The initial inventory data and associated model parameters are specified in Table 1.

A scenario-based approach is employed to investigate the release and transport of the C-14 associated with reactor graphite. In Scenario A, graphite is assumed to be segregated from the other wastes and packaged in concrete. Carbonation is assumed to dominate the behaviour of carbon dioxide and processes such as corrosion and microbial activity which may influence C-14 transport are considered to be negligible.

In Scenario B, the release of C-14 is modelled alongside the generation of hydrogen from the corrosion of steel packaging and waste components. Microbial activity is assumed to be

negligible due to the ambient pH of the repository and once again the behaviour of carbon dioxide is assumed to be dominated by carbonation.

In Scenario C, a biotic environment is considered and microbial activity is modelled, alongside corrosion and the release of C-14 from graphite. In this scenario two cases are examined. In the first case, C1, microbial activity is able to utilize carbon dioxide in the metabolism of corrosion hydrogen and the generation of methane with only excess carbon dioxide being removed by carbonation. In the second case, C2, all the carbon dioxide generated during cellulose degradation is removed by carbonation.

Results

The three scenarios considered generated different radiological dose profiles when modelled up to 10^5 y

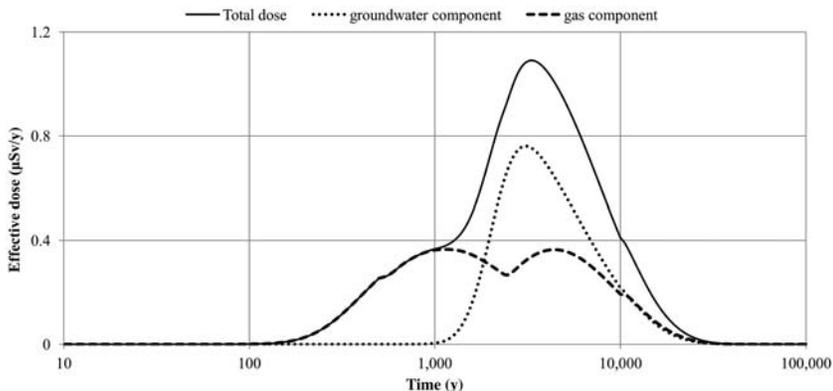


FIG. 4. Groundwater and gas components of the effective dose for scenario C1.

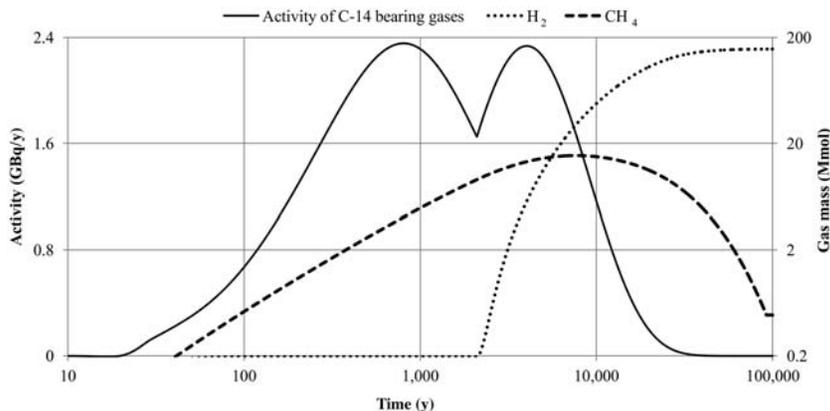


Fig. 5. Activity of C-14 bearing gases and biogenic gas mass for scenario C1.

(Fig. 3). In scenario A, the dose remains at $<0.01 \mu\text{Sv/y}$ with the groundwater release pathway dominating over the gaseous release pathway. The release of C-14 via the gas pathway is low in scenario A, due to the absence of a driving force associated with the generation of bulk gases by corrosion and organic degradation. Releases to the biosphere via the groundwater pathway are low due to the combined impacts of low near-field conductivity, carbonation, dispersion and radio-active decay.

Scenarios B and C2 have broadly similar dose profiles, the increased doses over and above those seen in scenario A are due to the generation of bulk gases which in turn drive the release of C-14 bearing gases. The doses are slightly higher in scenario C2, than those in scenario B; the difference being due to the larger gas volumes generated by microbial activity in scenario C2.

In scenario C1, the dose is significantly greater than that simulated in the other scenarios. This is largely due to the microbial reduction of C-14-bearing CO_2 to C-14-bearing CH_4 . This contrasts with the other scenarios where the majority of the C-14-bearing CO_2 is retained in the near field via carbonation reactions with cement.

The groundwater and the gas components of the effective dose for scenario C1, which are related with the groundwater pathway and the gas pathway, respectively, are shown in Fig. 4. The gas component increases earlier and is the only component of the dose for ~ 1000 y. After this time, the groundwater component also becomes an important contribution to the peak dose estimate. The double peak in the gas component is related to changes in the volumes of non radioactive gases generated in the near field and

driving the release of C-14 bearing gases. The start of the second peak represents the point at which corrosion-derived hydrogen begins to appear alongside microbially generated methane (Fig. 5). This occurs when the supply of CO_2 derived from organic-waste degradation is not sufficient to account for all the hydrogen generated by corrosion.

Discussion and conclusions

The modelling work reported here highlights a number of key processes that influence the radiological assessment of the disposal of reactor graphite and the associated C-14. The approach aligns with the opinions expressed by Bracke and Muller (2008) who argued for less conservative approaches to C-14 assessment modelling. The results are also in agreement with those of Towler *et al.* (2012) who took a different approach to represent a similar system. The modelling framework reported here could help to inform the possible segregation of reactor graphite from other gas-generating waste forms to minimize C-14 transport via the gas pathway.

The work also highlights the significance of the potential role of microbial reduction of C-14-bearing CO_2 to C-14 bearing CH_4 through the oxidation of corrosion hydrogen in increasing the mobility and possible radiological impacts of C-14. The importance of this process is being emphasized by experimental evidence for the release of C-14 from graphite as CO_2 (Baston *et al.*, 2014). Assessment and modelling studies have generally reduced the calculated impact of this process by invoking carbonation as a mechanism for the

diversion of C-14-bearing CO₂ into an immobilized and biologically inert form (Jackson and Yates, 2011; Towler *et al.*, 2012; Hoch *et al.*, 2008). The modelling results reported here indicate that microbial generation of C-14 bearing CH₄ is only significant when microbes have access to C-14 CO₂ prior to carbonation. In this case, the conversion of CO₂ to CH₄ becomes controlled by the availability of H₂ generated by corrosion. The importance of carbonation as an attenuating process for C-14 transport should also be viewed in the context of experimental evidence of the direct utilization of calcite by hydrogen oxidizing microbes under neutral and alkaline conditions (Suzuki *et al.*, 2014; Virden and Kral, 2010).

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