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1 **An approach to modelling the impact of ¹⁴C release from**
2 **reactor graphite in a geological disposal facility**

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7 **ABSTRACT**

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

12 There are a number of uncertainties associated with the fate and transport of
13 C-14 in a post-closure disposal environment that need to be considered when
14 calculating the radiological impacts of C-14 containing wastes. Some of these
15 uncertainties are associated with the distribution of C-14 containing gaseous
16 species such as ¹⁴CH₄ and ¹⁴CO₂ between the groundwater and gaseous release
17 pathways. As part of the C14-BIG programme, a modelling framework has been
18 developed to investigate these uncertainties. This framework consists of a
19 biogeochemical near-field evolution model, incorporating a graphite carbon-14
20 release model, which interfaces with a geosphere/biosphere model. The model
21 highlights the potential impact of the microbial reduction of ¹⁴CO₂ to ¹⁴CH₄,
22 through the oxidation of H₂, on C-14 transport. The modelling results could be
23 used to inform the possible segregation of reactor graphite from other gas
24 generating wastes.

25 **Keywords:** carbon-14, graphite, gas generation, geosphere, dose assessment

26 **Introduction**

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

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

39 The release of C-14 via a gas release pathway in a fractured higher strength
40 host rock is highly dependent on the volumes of non-radioactive gases generated
41 within a GDF, since these gases have the ability to drive the transport of the
42 smaller volumes of C-14 bearing gases. Microbiological and corrosion processes
43 may generate gas within a disposal facility and the associated biogeochemical
44 processes have been addressed in detail by several studies (Humphreys et al., 1997;
45 Graham et al., 2003; Suckling et al., 2011). The most mobile C-14 species is likely
46 to be CH₄ since this is not subject to precipitation reactions that will retard the
47 migration of C-14 labelled CO₂. If C-14 labelled CH₄ reaches the biosphere it will
48 be subjected to microbial oxidation to C-14 CO₂; once converted to CO₂ the C-14 is
49 available for root and foliar uptake by plants (Hoch et al., 2014).

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

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

68 As part of the C14-BIG programme, a modelling framework has been
69 developed that goes beyond that modelled by Towler et al. (2012) by integrating a
70 range of gas generation processes with recent insights into the speciation of C-14
71 leaching from reactor graphite (Baston et al., 2014). This framework consists of a
72 biogeochemical near-field evolution model, incorporating a graphite C-14 release

73 model, which interfaces with a geosphere/biosphere model developed in a GoldSim
74 (GoldSim, 2010) simulation environment.

75 **Model description**

76 *Near field Processes*

77 *C-14 release from reactor graphite*

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

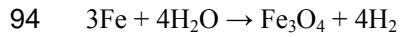
$$q = (k_{ca} F_{ca} e^{-k_{ca}t} + k_{cc} F_{cc} e^{-k_{cc}t}) A_0 e^{-\lambda_c t} \quad (1)$$

85 where q is the release rate of C-14 from graphite [TBq/a], k_{ca} and k_{cc} are the rate
86 constants for the rapid and the slow release [a^{-1}], respectively, F_{ca} and F_{cc} are the
87 rapidly and the slowly releasable fraction respectively, A_0 is the initial activity of
88 C-14 in the graphite [TBq], λ_c is the radioactive decay rate of C-14 [a^{-1}] and t is
89 time [a].

90 <Fig. 1>

91 *Corrosion*

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



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

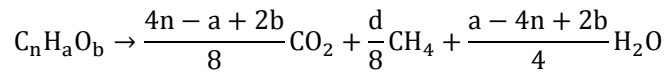
$$-\frac{dQ}{dt} = \frac{\rho r A}{M} \quad (2)$$

96 where Q is the mass of steel [mol], ρ is the density of steel [kg/m^3], M is effective
97 molecular mass of steel [kg/mol], r is the corrosion rate under anaerobic conditions
98 [m/a] and A is the surface area of steel [m^2].

99 *Microbial degradation*

100 Cellulose is the only class of organic waste considered within the model. The
101 microbial degradation of polymeric organics such as cellulose follows a generic
102 sequence of events (Rittmann and McCarty, 2001), where first the polymer is
103 converted into soluble intermediates. These are then subject to microbial oxidation
104 reactions depending on the prevailing geochemical conditions. The microbial
105 degradation of polymeric substrates and the subsequent generation of gas is a rather
106 complex process involving an array of microbial species. Representation of this
107 process can be simplified by the fact that the conversion of polymeric substrates to
108 their soluble intermediates is often the rate limiting factor in the gas generation
109 process (Rittmann and McCarty, 2001), allowing the microbial oxidation of
110 polymers to be coupled directly to the reduction of the relevant terminal electron
111 acceptors. This approach has been applied in other modelling studies (McNab and
112 Narasimham, 1994; Postima and Jacobsen, 1996; Suckling et al., 2011), since it

113 simplifies the model by reducing the number of kinetic parameters required. In the
 114 model reported here, organic degradation is modelled by the following reaction:



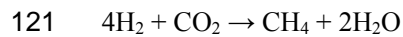
115 where $d=4n+a-2b$ (for cellulose: $n=6$, $a=10$, $b=5$, $d=24$).

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

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

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

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



122 The microbial consumption of hydrogen and the generation of hydrogen from
 123 corrosion have independent reaction rates. If the rate of microbial hydrogen
 124 metabolism is higher than that of corrosion and the associated hydrogen generation,
 125 then the rate of corrosion becomes the limiting process.

126 Microbes utilize part of the degraded organic material to produce new biomass
 127 of the composition $C_5H_7O_2N$. Microbes are also subject to natural turnover since
 128 cells have finite lifetimes and the model assumes that a fraction of the dead
 129 biomass is recycled to the organic material. The production rate of biomass is
 130 controlled by the degradation rate of the organic material and the decay rate of
 131 biomass, and is given by:

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

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

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

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

148 *Chemical Processes*

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

156 within the GDF (NDA, 2010a). Carbonation is important since it has the potential
157 to retain C-14 in the near-field through the precipitation of C-14 bearing CO₂.

158 *Additional aspects of near field modelling*

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

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

170 The gas generated in the near field is partitioned between the solution and the
171 gas phase using Henry's law. The gas phase is hosted in an assumed headspace
172 within the repository (Humphreys et al., 1997; Graham et al., 2003). The aqueous
173 phase is in equilibrium with the headspace gases and it is assumed that there is a
174 perfect mixing between the gases both in the headspace and the aqueous phase. The
175 carbonate chemistry associated with carbon dioxide solubility is not modelled
176 explicitly. Rather the carbonation process is modelled as a sink term for carbon
177 dioxide removal, an approach consistent with other gas generation models
178 (Rodwell 2004). A fractured higher strength host rock is assumed to host the
179 repository and as such is assumed not to represent a significant barrier to gas

180 migration (NDA, 2010c). Gas is released to the far field via the headspace based on
181 the mean near field gas pressure, which is determined by assuming that hydrostatic
182 conditions exist. This release of C-14 bearing gases to the far field constitutes the
183 gaseous release pathway.

184 *Far field modelling*

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

192 Fig. 2 shows the groundwater and gas pathway of C-14 from the repository to
193 the biosphere. Along the groundwater pathway, the model considers a constant
194 groundwater flow and advection and dispersion processes through a homogeneous
195 and isotropic geological medium. Groundwater abstraction is modelled from a near
196 surface aquifer system. The model allows dissolved C-14 species to be released
197 into the gas phase as the groundwater approaches the surface since water pressure
198 decreases along with associated solubility of C-14 bearing gases. Gaseous species
199 released from the groundwater are assumed to travel to the biosphere through faults
200 in the higher strength host rock following mostly a vertical elevation. The
201 migration of the free gas phase generated in the near field and in the groundwater

202 pathway is controlled by a constant gas flow rate depending on rock permeability,
203 flow area and pressure difference according to the Darcy flow equation.

204 <Fig. 2>

205 The biosphere environment is simulated by using three discrete model
206 compartments: the soil-plant, the animal uptake and the human uptake. The soil-
207 plant compartment is based on the enhanced RIMERS model (Thorne, 2005) and is
208 interfaced both with the gas and the groundwater pathway. Within the soil
209 compartment, $^{14}\text{CH}_4$ in the free gas phase is conservatively assumed to be fully
210 metabolised by microbes to $^{14}\text{CO}_2$. The animal and the human uptake
211 compartments are based on the ingestion rates of particular food sources and the C-
212 14 dose coefficients; these are only interfaced with the groundwater pathway. The
213 biosphere is modelled in line with the approach described by Limer et al. (2010).

214 *Scenarios description*

215 The modelling framework is applied to an assumed repository at a depth of
216 650 m containing intermediate level radioactive wastes based on the UK inventory.
217 Fully saturated and anaerobic conditions are assumed from the beginning of the
218 simulation since only the post-close environment is considered here. The initial
219 inventory data and associated model parameters are specified in Table 1.

220 <Table 1>

221 A scenario based approach is employed to investigate the release and transport
222 of the C-14 associated with reactor graphite. In Scenario A, graphite is assumed to
223 be segregated from the other wastes and packaged in concrete. Carbonation is

224 assumed to dominate the behaviour of carbon dioxide and processes such as
225 corrosion and microbial activity which may influence C-14 transport are
226 considered to be negligible.

227 In Scenario B, the release of C-14 is modelled alongside the generation of
228 hydrogen from the corrosion of steel packaging and waste components. Microbial
229 activity is assumed to be negligible due to the ambient pH of the repository and
230 once again the behaviour of carbon dioxide is assumed to be dominated by
231 carbonation.

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

239 **Results**

240 The three scenarios considered generated different radiological dose profiles
241 when modelled up to 10^5 years (Fig. 3). In scenario A, the dose remains below 0.01
242 $\mu\text{Sv/a}$ with the groundwater release pathway dominating over the gaseous release
243 pathway. The release of C-14 via the gas pathway is low in scenario A, due to the
244 absence of a driving force associated with the generation of bulk gases by
245 corrosion and organic degradation. Releases to the biosphere via the groundwater
246 pathway are low due to the combined impacts of low near-field conductivity,
247 carbonation, dispersion and radioactive decay.

271

<Fig. 5>

272 **Discussion and conclusions**

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

281 The work also highlights the significance of the potential role of microbial
282 reduction of C-14 bearing CO₂ to C-14 bearing CH₄ through the oxidation of
283 corrosion hydrogen in increasing the mobility and possible radiological impacts of
284 C-14. The importance of this process is being emphasised by experimental
285 evidence for the release of C-14 from graphite as CO₂ (Baston et al., 2014).
286 Assessment and modelling studies have generally reduced the calculated impact of
287 this process by invoking carbonation as a mechanism for the diversion of C-14
288 bearing CO₂ into an immobilised and biologically inert form (Jackson and Yates,
289 2011; Towler et al., 2012; Hoch et al., 2008). The modelling results reported here
290 indicate that microbial generation of C-14 bearing CH₄ is only significant when
291 microbes have access to C-14 CO₂ prior to carbonation. In this case, the conversion
292 of CO₂ to CH₄ becomes controlled by the availability of H₂ generated by corrosion.
293 The importance of carbonation as an attenuating process for C-14 transport should
294 also be viewed in the context of experimental evidence of the direct utilisation of

295 calcite by hydrogen oxidising microbes under neutral and alkaline conditions
296 (Suzuki et al., 2014; Virden and Kral, 2010).

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393

394

List of Tables

395 Table 1. Wastes in the assumed repository and the parameters of the modelling
 396 framework

Parameter	Value	Reference
Graphite radioactivity [TBq]	6000	Limer et al. (2010)
Cellulose mass [Mmol]	9.3	NDA (2010c)
Steel mass [Mmol]	1298	NDA (2010c)
Rapidly releasable fraction of C-14	$1.5 \cdot 10^{-4}$	Baston et al. (2014)
Slowly releasable fraction of C-14	$5 \cdot 10^{-2}$	Baston et al. (2014)
Fraction of C-14 released as CH ₄	10^{-2}	Baston et al. (2014)
Fraction of C-14 released as CO	$5 \cdot 10^{-3}$	Baston et al. (2014)
Rate of rapid release of C-14 [a ⁻¹]	50	Baston et al. (2014)
Rate of slow release of C-14 [a ⁻¹]	$2.5 \cdot 10^{-3}$	Baston et al. (2014)
Steel corrosion rate[$\mu\text{m/a}$]	10^{-2}	
Surface area of steel [km ²]	10	
Cellulose degradation rate [a ⁻¹]	$5 \cdot 10^{-4}$	Walke et al. (2011)
Hydrogen metabolism rate [a ⁻¹]	250	Walke et al. (2011)
Biomass yield coefficient [-]	0.23	Walke et al. (2011)
Biomass decay rate [a ⁻¹]	10	Walke et al. (2011)
Recycling fraction of dead biomass	0.9	Walke et al. (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]	$3.3 \cdot 10^{-12}$	
Hydraulic gradient of groundwater [-]	0.001	Limer et al. (2010)
Porosity in the near field [-]	0.1	
Porosity in the far field [-]	0.28	
Dispersivity [m]	10	
Diffusivity [m ² /s]	$4.3 \cdot 10^{-10}$	
Length of the groundwater pathway [m]	6000	
Release area of the gas pathway [m ²]	$18 \cdot 10^6$	
Flow area of the groundwater pathway [m ²]	20000	
Flow area of the gas pathway [m ²]	9	

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400 Fig. 1. Conceptual model of C-14 release from graphite (adapted from Baston et
401 al., 2014)

402 Fig. 2. Groundwater and gas pathways in the geosphere

403 Fig. 3. Radiological effective dose for the scenarios in the near field environment

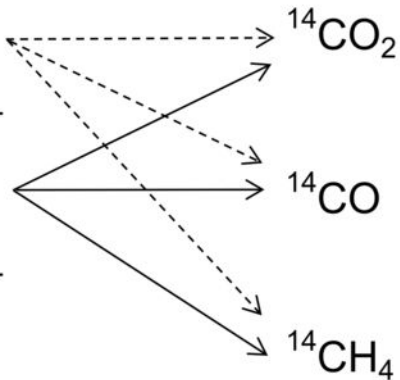
404 Fig. 4. Groundwater and gas components of the effective dose for scenario C1

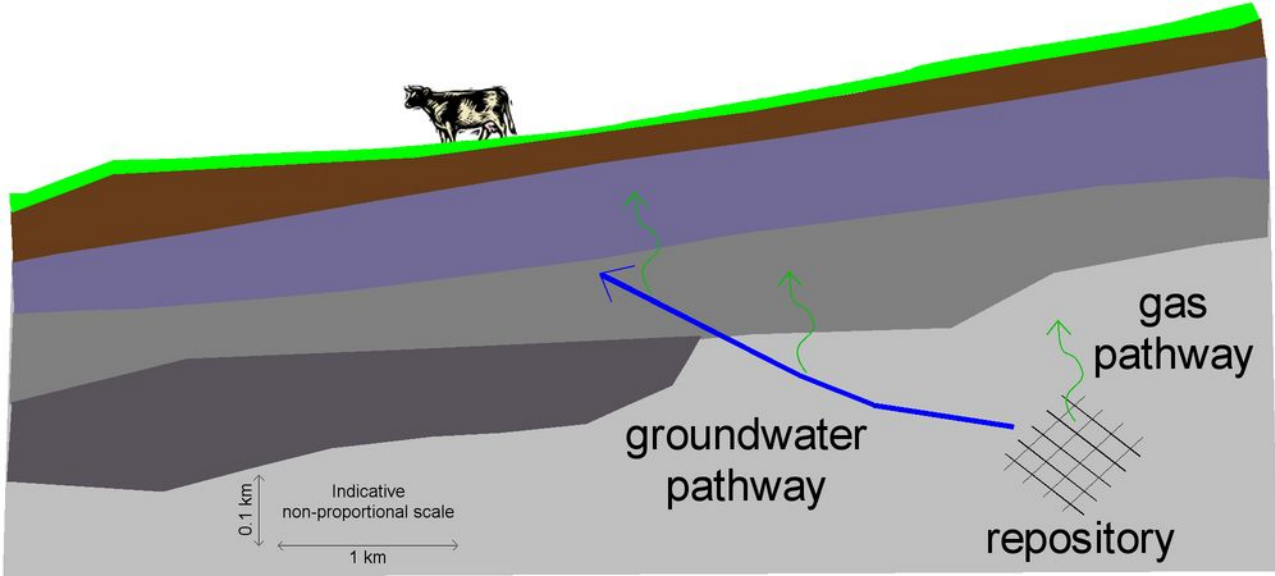
405 Fig. 5. Activity of C-14 bearing gases and biogenic gas mass for scenario C1

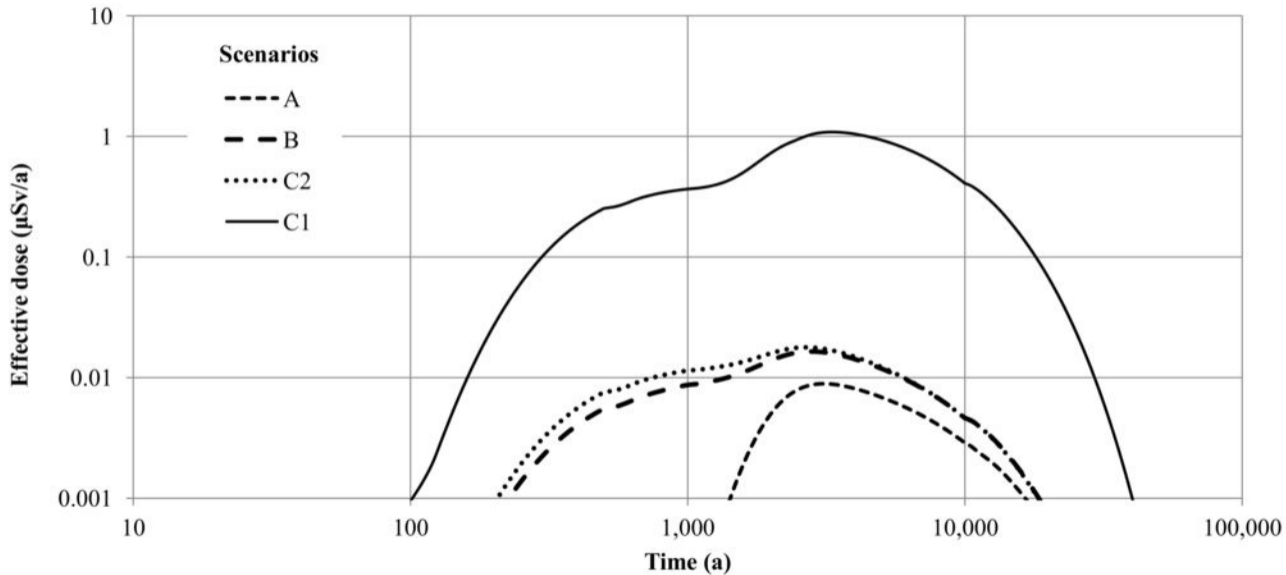
rapidly releasable
fraction of ^{14}C

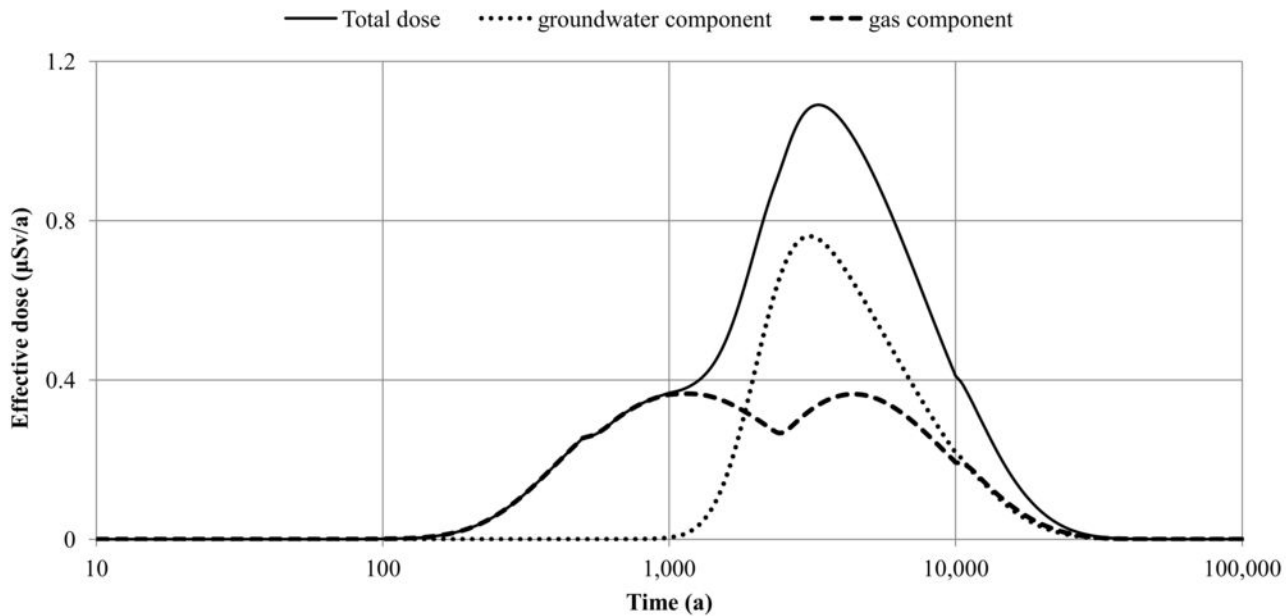
slowly releasable
fraction of ^{14}C

unreleasable
fraction of ^{14}C









— Activity of C-14 bearing gases H₂ - - - CH₄

