# PRELIMINARY INVESTIGATION OF $^{14}$C MIGRATION FROM RBMK-1500 REACTOR GRAPHITE DISPOSED OF IN A POTENTIAL GEOLOGICAL REPOSITORY IN CRYSTALLINE ROCKS IN LITHUANIA

Dalia Grigaliuniene[*] • Povilas Poskas • Raimondas Kilda • Asta Narkuniene

Nuclear Engineering Laboratory, Lithuanian Energy Institute, Breslaujos 3, 44403 Kaunas, Lithuania.

**ABSTRACT.** There are two units with RBMK-1500 type reactors at the Ignalina Nuclear Power Plant (Ignalina NPP) in Lithuania where graphite was used as a neutron moderator and reflector. These reactors are now being decommissioned, and Lithuania has to find a solution for safe irradiated graphite disposal. It cannot be disposed of in a near surface repository due to large amounts of $^{14}$C (radiocarbon, carbon-14); thus, a deep geological repository (DGR) is analyzed as an option. This study had the aim to evaluate $^{14}$C migration from the RBMK-1500 irradiated graphite disposed of in a potential DGR in crystalline rocks taking into account the outcomes of the research performed under the collaborative European project CAST (CArbon-14 Source Term) and to identify the potential to reduce the conservatism in the assumptions that was introduced in the lack of data and led in the overestimated $^{14}$C migration. The information gathered during the CAST project was used to model $^{14}$C transport in the near field by the water pathway and to perform uncertainty analysis. The study demonstrated that more realistic assumptions could reduce the estimated $^{14}$C flux from the near field by approximately one order of magnitude in comparison with the previous estimations based on very conservative assumptions.

**KEYWORDS:** deep geological repository, irradiated graphite, radionuclide $^{14}$C migration, uncertainty analysis.

## INTRODUCTION

The Ignalina Nuclear Power Plant (Ignalina NPP) in Lithuania has two RBMK-1500 type reactors and they are currently being decommissioned. Graphite in the RBMK-1500 type reactors was used as a neutron moderator and reflector. As a result of Ignalina NPP operation, about 3800 t of irradiated graphite (i-graphite) was generated (Poskas et al. 2012). One of the key radionuclides in the i-graphite is $^{14}$C. Its long half-life (5730 yr) and the role in biological processes require appropriate treatment and disposal of i-graphite waste. The number of initiated international projects, including the International Atomic Energy Agency (IAEA) Coordinated Research projects (Ojovan and Wickham 2014, 2016; Wickham et al. 2017) and collaborative European projects (Banford et al. 2008; Williams and Scourse 2015) clearly demonstrates the importance of this issue. The final decision on the i-graphite management route in Lithuania has not yet been taken; however, disposal in a deep geological repository (DGR) is considered as a potential solution.

Up to now, the research of RBMK-1500 i-graphite disposal in Lithuania has been limited. Initial analysis of possibilities of disposing of i-graphite in a DGR in crystalline rock was presented in (Narkuniene et al. 2014). Two alternatives were analyzed: i-graphite disposal in metal containers without an encapsulant and disposal of encapsulated waste. The focus of the work was mainly on the evaluation of the importance of the waste leaching rate to the $^{14}$C transfer into the geosphere and the potential radiological impact. An insufficient amount of modeling data on the source term determined that the evaluation was performed with very conservative assumptions.

*Corresponding author. Email: Dalia.Grigaliuniene@lei.lt.

The next step in the investigations related to i-graphite disposal in a DGR in Lithuania was identification of the most important near field parameters governing the release and transport of [14]C (Poskas et al. 2016). For this purpose, the assessment of ^{14}C transfer through engineered barriers into the geosphere was reassessed using the information on the source term which was available at that time and a local sensitivity analysis was performed. The assessment revealed that for inorganic ^{14}C compounds, depending on the alternative, the maximal fractional flux into the geosphere (i.e. the ratio of the released activity and the total disposed activity) can vary from $10^{-11}$ yr$^{-1}$ to $10^{-12}$ yr$^{-1}$, while for organic ^{14}C compounds, it is about $10^{-3}$ yr$^{-1}$. Based on results of the local sensitivity analysis, it was concluded that parameters with the highest influence on the fractional flux into the geosphere are the distribution coefficient indicating ^{14}C sorption in the backfill and the backfill hydraulic conductivity.

In the course of the EC 7FP collaborative project CAST (Carbon-14 Source Term) (2013-2018) (CAST 2013) investigations into ^{14}C inventory in i-graphite, its release and speciation have been performed. The outcomes of these investigations together with the most recent results from the research carried out under national UK and French programs have formed the basis for a more realistic modeling of the potential release and migration of the ^{14}C in the near field. The aim of this paper was to incorporate the information gathered during the CAST project in the context of the previous assessment of the ^{14}C migration from the RBMK-1500 i-graphite disposed of in a DGR in crystalline rocks and to identify the areas for conservatism reduction.

## METHODOLOGY

The estimation of the ^{14}C migration from the i-graphite disposed of in a DGR is performed following the structure of the safety assessment methodology presented in the IAEA Specific Safety Guide SSG-23 (IAEA 2012). Following this structure, a framework for the assessment was defined, and the near field (engineered barrier system) was specified. A scenario of the potential ^{14}C release from i-graphite and transfer in the near field was then developed and transformed into the conceptual model amenable to mathematical representation. Solution of the mathematical model was achieved by implementing the model into a software tool. Finally, calculations were undertaken and analyses and interpretation of the results were performed. When modeling ^{14}C migration, at first deterministic calculations were carried out, assigning probable realistic values to the parameters. This case is further referred as the Base Case. The obtained results were compared with the results of the previous conservative assessment presented in (Poskas et al. 2016). Probabilistic calculations were then performed, in order to identify how the uncertainties in the parameter values could influence the modeled ^{14}C flux from the near field.

## Assessment Context

The Republic of Lithuania is a Member State of the European Union (EU) and is obliged to implement Council Directive 2011/70/EURATOM that establishes a Community framework for the responsible and safe management of spent nuclear fuel (SNF) and other radioactive waste in the EU (EU 2011). In 2015, in line with the provisions of this document, the Government of the Republic of Lithuania approved a National Radioactive Waste Management Development Program (LR 2015), which states that storage of SNF and long-lived radioactive waste (to which i-graphite is also attributed) is solely a temporary decision, and disposal of SNF and long-lived radioactive waste in a geological repository is the only sustainable method of the final disposal. Following the provisions indicated in the Program, the possibility to dispose of the i-graphite from the Ignalina NPP in a DGR in crystalline rock is investigated in this study. At this stage of the repository development the investigations of the ^{14}C migration are limited to the near field

environment. The modeling of $^{14}$C migration from the RBMK-1500 i-graphite is based on the outcomes of the research performed under the CAST project and national programs.

**Repository Concept**

Lithuania is at an early stage of a DGR development program, and there has been no decision on the DGR disposal site yet. It was identified from the available information about geological formations that a potential location for the DGR could be in the eastern part of Lithuania, where crystalline rock is at the depth of about 700 m. Granitic rock is covered by sedimentary rocks of different hydrogeological properties forming aquifers and aquitards. For $^{14}$C release and migration from the RBMK-1500 i-graphite disposed of in a DGR, the disposal concept was adopted from the assessment reported in (Poskas et al. 2016). It was assumed that i-graphite is disposed of in metal containers stacked in a separate tunnel of a DGR, see Figure 1a. Two options for i-graphite disposal were analyzed: in Alternative 1, it was assumed that the graphite waste inside the metallic containers is without an encapsulant, and in Alternative 2, waste encapsulated in the metallic containers with a cement-based material was considered. At the closure of the repository, backfilling of the tunnel with a cementitious material was assumed.

**Scenario Description**

The review of available experimental results indicated that majority of $^{14}$C release from i-graphite occurs in solution and only small amounts of gaseous releases (about 1% of the released $^{14}$C) have been measured (Toulhout et al. 2015). Estimation of the hydrogen gas generation rate due to container corrosion and transport in the near field revealed that formation of the bulk gas is hardly expected. Therefore, at this stage of the DGR development, the assessment of release and migration of the $^{14}$C from the RBMK-1500 i-graphite in the near field considers $^{14}$C transfer by the water pathway. A reference scenario is analyzed, i.e. no disruptive events are taken into account. The assessment is performed for the period after the repository closure assuming fully water saturated conditions from the beginning of the assessment. Upon contact with the water, $^{14}$C is leached from the i-graphite. In the absence of the site characterization data it is assumed that a fracture is intersecting the repository and leached $^{14}$C is transported to this fracture.

**Conceptual Model**

The conceptual model for the developed scenario is presented in Figure 1b. The following processes are considered: $^{14}$C leaching from the i-graphite, advective and diffusive transport
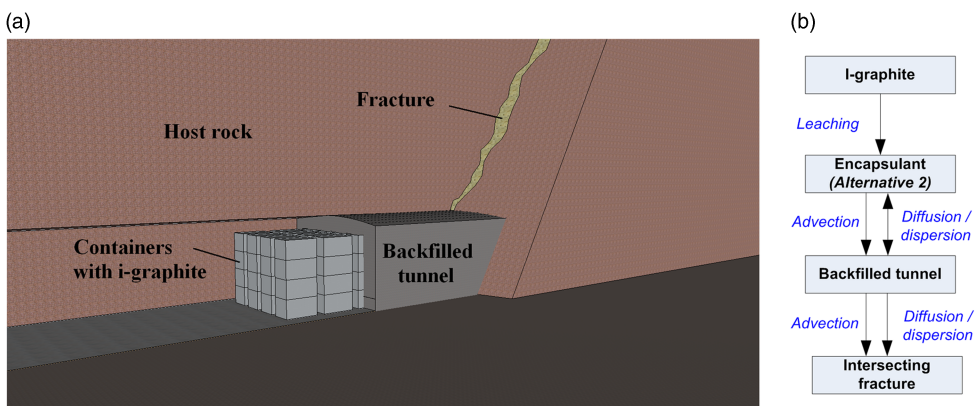


Figure 1    Repository concept (a) and conceptual model of $^{14}$C migration (b).

through the encapsulant (relevant only for Alternative 2) and the backfill and radioactive decay. There are indications that leaching of the cement under deep geological conditions is very slow (Wang et. al. 2012) and for the preliminary assessment degradation of the cement based materials is not modeled.

## Source Term

*Inventory.* $^{14}$C inventory in i-graphite depends on the location in the reactor core, operating power history, initial concentration of impurities, amount of cooling gases, etc. (Toulhout et al. 2015). In the previous assessments, the inventory was selected based either on the activation modeling with very conservative assumption on N impurity (Narkuniene et al. 2014) or on activity measurements at a single location in the reactor core (Poskas et al. 2016).

During the CAST project, a new numerical model for neutron activation was developed, and activity distribution of the $^{14}$C within the whole reactor graphite stack at the Ignalina NPP was obtained. The measured $^{14}$C content in the Ignalina NPP reactor GR-280 grade graphite reported in (Mazeika et al. 2015) is $1.67 \times 10^5$ Bq g$^{-1}$. Combining this data with the new model, estimation of the $^{14}$C inventory in the graphite stack was made (Narkunas and Poskas 2017). It was reported that the average specific activity value of the $^{14}$C in RBMK-1500 i-graphite was $1.9 \times 10^5$ Bq g$^{-1}$ with the standard deviation of 36.7%. These values were employed in this study for the assessment of the $^{14}$C release and migration.

*Leaching.* Experimental results indicate that $^{14}$C from i-graphite is released initially at a higher release rate followed by a decreased release rate in the long-term. However, for the treated graphite, the initial rapid release is not identified. The release rate also depends on the geometry of the sample: a higher release rate for crushed graphite is observed (Toulhout et al. 2015). Based on such information, in the previous $^{14}$C release assessments (Narkuniene et al. 2014; Poskas et al. 2016), several deterministic calculations were performed. The cases with and without rapid release as well as the bounding case with instant release of the total inventory were included. To represent rapid release, for the first 10 years after repository closure, fractional release rate of 0.1 yr$^{-1}$ was assumed, while the long-term fractional release rate was varied from $1.83 \times 10^{-5}$ yr$^{-1}$ to 0.1 yr$^{-1}$.

In the course of the CAST project, some more information regarding the release rate from i-graphite was reported, mainly as part of the UK program (AMEC 2016). First of all, leaching experiments with the UK graphite provided evidence that a significant proportion of the $^{14}$C content is unleachable. According to AMEC (2016), under conditions expected in a DGR, the releasable $^{14}$C makes between about 1% and 5% of the total $^{14}$C inventory and even under harsh acidic conditions not more than 30% of the $^{14}$C is released. These findings were used for the parameterization of the revised UK model of the $^{14}$C release. It was assumed that the rapid release fraction can vary in the interval from 0 to $2 \times 10^{-3}$ with the best estimate value of $2 \times 10^{-4}$; the lower bound of the fraction of the $^{14}$C that is available for slower release is 0.01, the upper bound 0.3 and the best estimate value 0.05 (AMEC 2016). Rate constant for the slower release of $^{14}$C provided in (AMEC 2016) varies from $10^{-3}$ yr$^{-1}$ to 0.1 yr$^{-1}$ with the best estimate value of $10^{-2}$ yr$^{-1}$.

As the research performed under the UK program was carried out for other type i-graphite than RBMK-1500, the presented above UK findings are cautiously used in the $^{14}$C release assessment in the current study by assuming that the releasable fraction makes 0.3 from the total $^{14}$C inventory. The fraction of the rapid release was assumed the same as in the

UK model and based on Toulhout et al. (2015) and AMEC (2016) was considered as instant release. The rest from the releasable inventory was associated with slower long-term release with the release rate as in the UK model. The selected long-term release rate is conservative in comparison with $^{14}C$ release rate from French and Japanese graphite (Toulhout et al. 2015).

*Speciation.* Released from i-graphite, $^{14}C$ can form both organic and inorganic compounds. It is expected that for the conditions prevailing in a DGR after its closure and for slow degradation of i-graphite, mainly $^{14}CO_2$/carbonate and $^{14}CH_4$ will be formed (Toulhout et al. 2015).

Due to insufficient data on partitioning of the released $^{14}C$ between organic and inorganic compounds, in the previous assessments (Narkuniene et al. 2014; Poskas et al. 2016), two cases were analyzed: in one case it was assumed no sorption of the released $^{14}C$ in engineered barriers, and in another case strong sorption in cementitious material was assumed.

There is still no clear picture on partition of the released $^{14}C$ between organic and inorganic compounds. On the one hand, experiments on $^{14}C$ speciation under modeled DGR conditions performed in France revealed that the fraction of inorganic $^{14}C$ was from 65% to 75% with the rest attributed to organic compounds (Toulhout et al. 2015). On the other hand, from the leaching studies of moderator and reflector graphite from the Tokai (Magnox) reactors in Japan it was found that about 80% of the released $^{14}C$ was in organic form (AMEC 2016). Work undertaken as part of the UK program identified that under anaerobic high pH conditions (as expected in the DGR in the long term) released $^{14}CO_2$ fraction makes 0.99 (AMEC 2016).

Taking into account that there are no experimental studies on $^{14}C$ compounds released from RBMK-1500 i-graphite and available results on partition between organic and inorganic compounds from research with other types of i-graphite vary in a large interval, for the uncertainty analysis in the current study it was assumed that the fraction of organic compounds in solution vary between 20% and 80% with the best estimate value of 50%. The remaining fraction is assigned to inorganic compounds. Table 1 shows the source term parameter values from previous assessments and the values assumed in the present study.

**Mathematical Model**

$^{14}C$ transfer in the near field is described by an advection-dispersion equation, which in one-dimensional form is:

$$\frac{\partial}{\partial t}(R\,\theta\,C) = \frac{\partial}{\partial x}\left(\theta D \frac{\partial C}{\partial x}\right) - \frac{\partial}{\partial x}(qC) - \lambda\theta\,RC,$$

where $C$ is $^{14}C$ concentration in water (Bq m$^{-3}$), $\theta$ is effective porosity (–), $R$ is retardation coefficient (–), expressed as $R = 1 + \rho \times K_d / \theta$, $\rho$ is bulk density (kg m$^{-3}$), $K_d$ is sorption coefficient (m$^3$ kg$^{-1}$), $D$ is diffusion-dispersion coefficient (m$^2$ s$^{-1}$), expressed as $D = D_{eff} + \alpha \times q / \theta$, $D_{eff}$ is effective diffusion coefficient (m$^2$ s$^{-1}$), $\alpha$ is longitudinal dispersivity (m), $q$ is water flow rate (m s$^{-1}$), $\lambda$ is radioactive decay constant (s$^{-1}$), $x$ is distance in the direction of water flow (m), $t$ is time (s). The initial condition used in the model is that there is no $^{14}C$ present at t = 0 around waste matrix and the boundary condition at the output to the fracture has been implemented assuming zero concentration outside the boundary.

The near field model was implemented in the software tool AMBER (Quintessa 2012). For the modeling purposes, a single central container with external dimensions of

Table 1   Comparison of the source term parameter values assumed in the previous assessments and in the present study.

| Parameter | Narkuniene et al. (2014) | Poskas et al. (2016) | Present study |
|---|---|---|---|
| Inventory, Bq g$^{-1}$ | $9.9 \times 10^5$ | $1.67 \times 10^5 \pm 36.7\%$ | $1.9 \times 10^5 \pm 36.7\%$ |
| Releasable fraction ($f$), – | 1 | 1 | 0.3 |
| Rapid release fraction ($f_r$), – | 0 to 1 | 0 to 1; best estimate 10% from remaining amount for 10 years | 0 to $2 \times 10^{-3}$; best estimate $2 \times 10^{-4}$ |
| Rapid release rate, yr$^{-1}$ | 0.1 to Instant | 0.1 | Instant |
| Slower release fraction ($f_s$), – | $1 - f_r$ | $1 - f_r$ | $f - f_r$ |
| Slower release rate, yr$^{-1}$ | $1.83 \times 10^{-5}$ to 0.1 | $1.83 \times 10^{-5}$ to 0.1; best estimate $10^{-3}$ | $10^{-3}$ to 0.1; best estimate $10^{-2}$ |
| Organic $^{14}$C fraction ($f_{org}$), – | 0 or 1 | 0 or 1 | 0.2 to 0.8; best estimate 0.5 |

4.013 m $\times$ 2.438 m $\times$ 2.2 m in the tunnel was considered assuming that the average distance from the point of release of $^{14}$C to reach the intersecting fracture is 25 m (based on Towler et al. 2010). It should be noted that the presence of the fracture is not definite and should be regarded as a conservative assumption in this study. I-graphite inside the container was modeled as a homogenous block. In Alternative 2, the thickness of the encapsulant surrounding the i-graphite was assumed to be 0.15 m. Properties of the tunnel backfill and the i-graphite encapsulant were selected based on Towler et al. (2010) and Limer et al. (2010). Modeling was performed with the backfill bulk density of 1730 kg m$^{-3}$, hydraulic conductivity $6 \times 10^{-8}$ m s$^{-1}$ and porosity 0.55. For the encapsulant, the bulk density of 2100 kg m$^{-3}$, hydraulic conductivity $10^{-9}$ m s$^{-1}$ and porosity 0.125 were assumed. Effective diffusion coefficient in the backfill and in the encapsulant was taken to be $10^{-11}$ m$^2$/s$^{-1}$ (based on IAEA 2004). The hydraulic gradient for the groundwater flow was 1%, longitudinal dispersivity – 1/10 of the path length.

When modeling $^{14}$C migration through engineered barriers, it was assumed in the previous assessments (Narkuniene et al. 2014; Poskas et al. 2016) that non-sorbed $^{14}$C released from i-graphite had no interaction with the cementitious backfill and the encapsulant and migrated without retardation. No sorption of organic compounds was also assumed in a number of works collected in the CAST project report (Kendall et al. 2015). During the CAST project it was obtained that sorption of small organic molecules in cement may be non-negligible, i.e. there are some experiments indicating sorption coefficient $K_d$ values in the interval from $10^{-3}$ m$^3$ kg$^{-1}$ to $10^{-5}$ m$^3$ kg$^{-1}$ with the best estimate value of $10^{-4}$ m$^3$ kg$^{-1}$ (Capouet et al. 2017). However, the dataset on $K_d$ values for organic $^{14}$C is rather limited therefore, in this study for the Base Case estimation it is assumed that organic compounds released from i-graphite migrate without retention and for the uncertainty analysis, the range of $K_d$ values from 0 to $10^{-4}$ m$^3$ kg$^{-1}$ was defined. $K_d$ value for inorganic compounds conservatively was taken to be 0.2 m$^3$ kg$^{-1}$ (based on Towler et al. 2010) with $\pm$ one order of magnitude variation for the uncertainty analysis and solubility limitation was not considered.
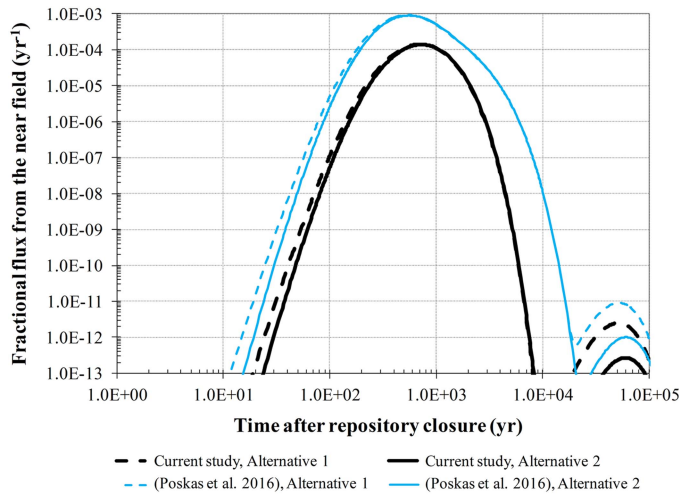
Figure 2   $^{14}$C fractional flux from the near field: Alternative 1 – non-encapsulated waste; Alternative 2 – encapsulated waste.

## RESULTS AND DISCUSSION

### Base Case

The fractional flux (estimated flux Bq yr$^{-1}$ per Bq of $^{14}$C disposed of in the repository) from the near field to the intersecting fracture is considered as the output from the modeling of the $^{14}$C release from the RBMK i-graphite and migration in the near field. The Base Case modeling results are presented in Figure 2. In addition, the results of the assessment performed by Poskas et al. (2016) are presented. The first peak in Figure 2 corresponds to the organic $^{14}$C flux from the near field and the second one to the inorganic $^{14}$C flux.

Looking at the results obtained in this study, it can be observed that the maximal fractional flux of the $^{14}$C released from the near field as organic compounds in both alternatives is approximately $1 \times 10^{-4}$ yr$^{-1}$, and the time needed to reach the maximum is about 500 years after the repository closure. The difference of the organic $^{14}$C maximal fractional flux from the near field between Alternative 1 (non-encapsulated waste) and Alternative 2 (encapsulated waste) makes about 2%. Such a small difference suggests that under assumptions introduced in the assessment, the additional cementitious barrier (encapsulant) play insignificant role in reducing the organic $^{14}$C flux from the repository.

A different result is observed for inorganic $^{14}$C. The maximal fractional flux of inorganic $^{14}$C in Alternative 1 is approximately $5 \times 10^{-12}$ yr$^{-1}$. When it is assumed that the i-graphite is encapsulated in a cementitious material, the maximal fractional flux is reduced by about one order of magnitude. The maximal inorganic $^{14}$C flux from the near field appears in about 50,000 years after the repository closure. Late inorganic $^{14}$C release from the near field and significantly reduced maximal fractional flux in Alternative 2 are mainly the result of good retention in the cementitious barriers.

The comparison of the Base Case results from the present study with the results reported in (Poskas et al. 2016) indicates that the maximal fractional flux from the near field in the present study is lower by about one order of magnitude. The results demonstrate that more realistic assumptions could significantly reduce the conservatism.
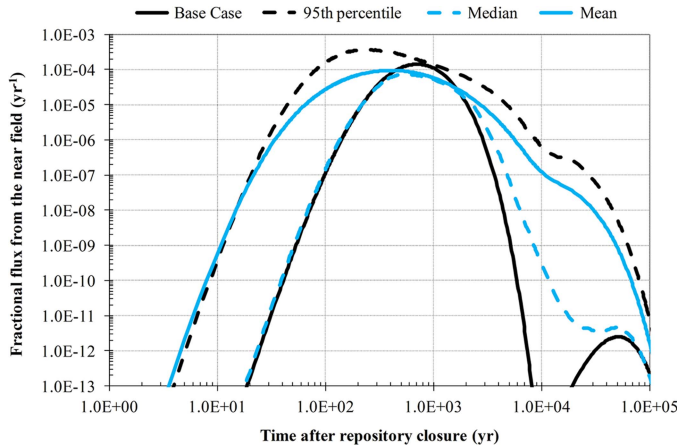
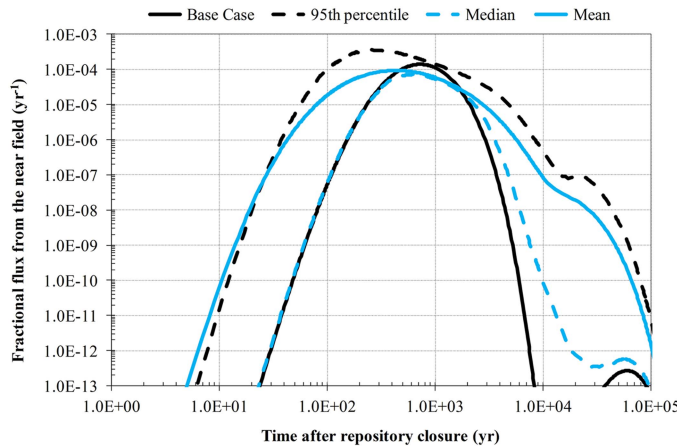Figure 3   Results of the uncertainty analysis, Alternative 1 (non-encapsulated waste).



Figure 4   Results of the uncertainty analysis, Alternative 2 (encapsulated waste).

## Uncertainty Analysis

For the uncertainty analysis, 2000 sets of parameter values were randomly generated from the respective probability density functions applying Monte-Carlo technique and calculations were performed. Results of the uncertainty analysis for Alternative 1 (non-encapsulated waste) are presented in Figure 3 and for Alternative 2 (encapsulated waste) in Figure 4. They provide plots showing the fractional $^{14}$C flux from the near field for the Base Case, the mean fractional flux and the 95th percentile. In addition, the median fractional flux value is added as it might be considered as a better indication of the central tendency.

It can be seen from Figure 3 that the maximal fractional flux values corresponding to the organic $^{14}$C release differ between the median value and the 95th percentile by about factor of 5. The maximal fractional flux from the near field for the Base Case calculations is about $1.4 \times 10^{-4}$ yr$^{-1}$, the maximum of the 95th percentile is about $3.7 \times 10^{-4}$ yr$^{-1}$, and the maximum of the median flux is about $7.2 \times 10^{-5}$ yr$^{-1}$.

The maximal fractional flux corresponding to inorganic $^{14}$C release varies in a much wider interval than in the case of organic $^{14}$C, i.e. from $2.5 \times 10^{-12}$ yr$^{-1}$ (Base Case) to $3.2 \times 10^{-7}$ yr$^{-1}$ (the 95th percentile). It is supposed that this is related to the changes in sorption coefficient values, as it was reported in (Poskas et al. 2016) that for inorganic $^{14}$C, sorption in cementitious barriers has the highest impact on $^{14}$C flux.

The results for Alternative 2 (see Figure 4) were obtained very similar to the results of Alternative 1 in the flux profile, with about one order of magnitude lower peak corresponding to inorganic $^{14}$C.

From the uncertainty analysis it can be concluded that further investigations in partitioning of the released $^{14}$C between organic and inorganic compounds and sorption of the released compounds in the cementitious environment could reduce the uncertainty of the results and provide a more realistic picture of the system's capability to provide the adequate level of safety.

## CONCLUSIONS

The outcomes of the research performed in the frame of the CAST project and information provided from the national research programs on $^{14}$C inventory in i-graphite, rapid and slow release fractions and release rate as well as on speciation and sorption of $^{14}$C in a cementitious environment were analyzed, and an updated set of parameter values was compiled for the modeling of $^{14}$C release from RBMK-1500 i-graphite disposed of in a DGR and for $^{14}$C migration through the engineered barriers. It was found that the information obtained during the CAST project incorporated in the assessment reduced the conservatism in the assumptions, and the estimated $^{14}$C flux from the near field decreased by about one order of magnitude in comparison with the previous conservative estimations.

The results of the uncertainty analysis demonstrated that further investigations in partitioning of released $^{14}$C between organic and inorganic compounds and sorption of the released compounds in a cementitious environment could further reduce the uncertainty in the modeling results.

However, the information on $^{14}$C release and speciation obtained during the CAST project was for other types of i-graphite than the RBMK-1500 reactor. In order to increase confidence in the justification of the parameter values and to get a more realistic representation of the system, experimental investigations into $^{14}$C release from RBMK-1500 i-graphite and speciation are still needed.

## ACKNOWLEDGMENTS

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