

## EVALUATION OF THE RADIOLOGICAL CONSEQUENCES OF $^{14}\text{C}$ DUE TO CONTAMINATED IGNALINA NPP GRAPHITE INCINERATION\*

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After decommissioning of Unit 1 of the Ignalina Nuclear Power Plant, the problem of the radioactive waste management emerged. Among radioactive waste there is an inventory of about 1700 tons of the graphite containing  $^{14}\text{C}$  radioisotope as an activation product. The estimates show that the maximal total inventory of  $^{14}\text{C}$  in graphite from Unit 1 is around  $7 \cdot 10^{14}$  Bq. One of the possible ways for utilization of the graphite is its incineration in the radioactive waste processing plant. Unfortunately, in this case a significant amount of the radionuclide would be released into the atmosphere in the form of  $\text{CO}_2$  and the released radiocarbon would cause additional exposure of the population. Possible radiological consequences for the Lithuanian inhabitants are evaluated using the model of radiocarbon dispersion in the environment and considering several scenarios of the graphite incineration. Dispersion of the incineration gas is modelled using the Gaussian dispersion model. Assimilation of  $\text{CO}_2$  by the vegetation due to photosynthesis as well as washout of  $\text{CO}_2$  from the atmosphere by rain, uptake of the deposited  $^{14}\text{C}$  by the plants from soil, and the eventual contamination of food products are considered. An estimated additional exposure effective dose to the critical group of the local population due to continuous releases of the total inventory of  $^{14}\text{C}$  from the incinerator is of the order of 2.7 mSv. The consumption of the contaminated locally produced food products is the main contribution to the dose. Such continuous incineration of graphite would be acceptable if it were extended for at least 14 years in order not to exceed the annual dose limit of  $0.2 \text{ mSv} \cdot \text{y}^{-1}$ . The incineration of graphite would cause the least radiological consequences if it functioned only in the dark time of the day or in winter when plants do not perform photosynthesis. In this case the effective dose for the population would be of the order of  $5.2 \mu\text{Sv}$ . World population would receive an average lifetime ( $\sim 50$  years) dose of  $0.43 \mu\text{Sv}$  per person which is negligibly small.

**Keywords:** radioactive waste management, graphite incineration,  $^{14}\text{C}$ , modelling, exposure doses

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### 1. Introduction

Unit 1 of the Ignalina Nuclear Power Plant (INPP) was shut down on 31 December 2004. Unit 2 is planned to be shut down in 2009. It is estimated that decommissioning of the plant will generate approximately 68000 tons of the radioactive waste of various levels of activity and will contain different radionuclides [1]. An optimal strategy needs to be found for the utilization of such a huge amount of the radioactive waste.

Significant part of the radioactive waste is graphite, which was used as a neutron moderator and reflector in the active zone of the reactor. Graphite accumulates significant amount of  $^{14}\text{C}$  and some other radio-

nuclides due to activation processes during operation of the plant. Inventory of the graphite is  $\sim 1700$  tons in each INPP reactor unit.

For the safe utilization of the graphite several strategies can be considered. One of the possibilities is the graphite storage in an intermediate radioactive waste repository. Such practices were currently applied in France but only negligible part of all graphite could be stored in such a repository due to its limited capacity. Intermediate depth or deep geological repositories (not in operation yet) could be considered as well. However, they have several disadvantages too. Graphite is a flammable material. Storage of graphite for a long period of time gives rise to a considerable degree of risk of the fire accidents. In the case of the accident, uncontrolled release of radionuclides into

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the environment would occur. In addition, storage and maintenance of the large volumes of the contaminated graphite in the depository would demand great expenses. Therefore, controlled incineration of the Ignalina NPP graphite in the specialized facility can be considered as an appropriate alternative instead of its disposal in a repository. Radiological impact of the incineration of the contaminated graphite was studied by several authors [2–4]. These studies showed that in principle such a way of the graphite utilization can be used if precautions are taken during release (stack height, daytime or night time conditions, etc.). However, radiological consequences from every incinerator have to be examined in detail in order to take into account the site specific conditions. Operation of the incinerator would be acceptable if radiation safety criteria were met for the most exposed population group.

## 2. Assessment of the radiological consequences

The fluidized bed incinerator of the activated graphite with application of the filters permit capturing the radionuclides contained in graphite, except for the releases of tritium and  $^{14}\text{C}$ , the latter being converted to the form of  $\text{CO}_2$ . Radiological impact of tritium is much lower than that due to  $^{14}\text{C}$ , therefore only radiocarbon is considered in the further analysis. Released  $\text{CO}_2$  participates in the turnover of carbon in the life cycle of the vegetation and animals. Because of the long half-life of  $^{14}\text{C}$  ( $T_{1/2} = 5730$  years) its radiological consequences for world population must be evaluated through the collective effective dose commitment, which is complete after about 50 000 years following the release.

Radiological impact on the population residing in the vicinity of the incinerator depends on several factors: amount and composition of the release from the incinerator; dispersion and retention of the released radionuclides in the environment. Influence of various conditions during incineration, such as time of the day during release, seasonality, and weather conditions, should be considered in order to select the optimal strategy for the graphite incineration. It should ensure that minimal radiological impact on the population would take place.

## 3. Activation analysis of graphite in the Ignalina NPP

Two codes of numerical calculations have been used for the determination of the inventory and concentra-

tion of radiocarbon and other radionuclides important from the radiological point of view in the used graphite of the Ignalina NPP Unit 1 reactor. The code MCNP for the estimation of the neutron transport is based on the Monte Carlo method, and the code CINDER90 uses the deterministic approach to calculate the activation of materials. Both codes are elaborated by Los Alamos National Laboratory (U.S.A.) and have been validated for the numerous calculations of the critical systems and activation of its constructions. The scheme used for qualitative and quantitative investigations of neutron flux and the activation of the graphite constructions of the RBMK reactor is given in Fig. 1.

The geometry of constructions of the RBMK reactor and their materials are described in 3D using conventional input cards of the MCNP code. To estimate the average flux of neutrons the burnup of 10 MWd/kgU is used. The fuel channels, fuel assemblies, control rods, and the prisms of graphite are represented as detailed as possible. MCNP estimates the events that appear during the neutrons transport in the material (leakage of the neutrons from the system, scattering, capture, fission) with their probability given in the nuclear data libraries, such as ENDF, JENDL, or JEF. Normalization of these estimates by the thermal power of the reactor gives the neutron fluxes in the constructions concerned. Such fluxes are further used by the code CINDER90 for the activation calculations. CINDER90 also uses nuclear data, the power history of the reactor, and the impurities in graphite, and solves the equations of nuclear transmutation and of radioactive decay for each radionuclide chain. Finally, the specific activities or radiotoxicities of radionuclides are obtained [5]. Table 1 summarizes the main radionuclides that occur in graphite of the Ignalina NPP RBMK reactor after 21 years of its operation.

The impurities in the fresh graphite sample of the Ignalina NPP reactor have been experimentally obtained by the irradiation-activation technique and by glow discharge mass spectrometry methods in C.E.A. Saclay scientific centre of the Commission for Atomic Energy (France). We have estimated that the maximal total inventory of  $^{14}\text{C}$  in the used graphite from the INPP Unit 1 reactor is  $7 \cdot 10^{14}$  Bq. It is the upper limit estimate, derived assuming that activation products are retained in the graphite.

## 4. Radiological impact on the local population

After carbon is released into the environment, it is distributed among various compartments of the global

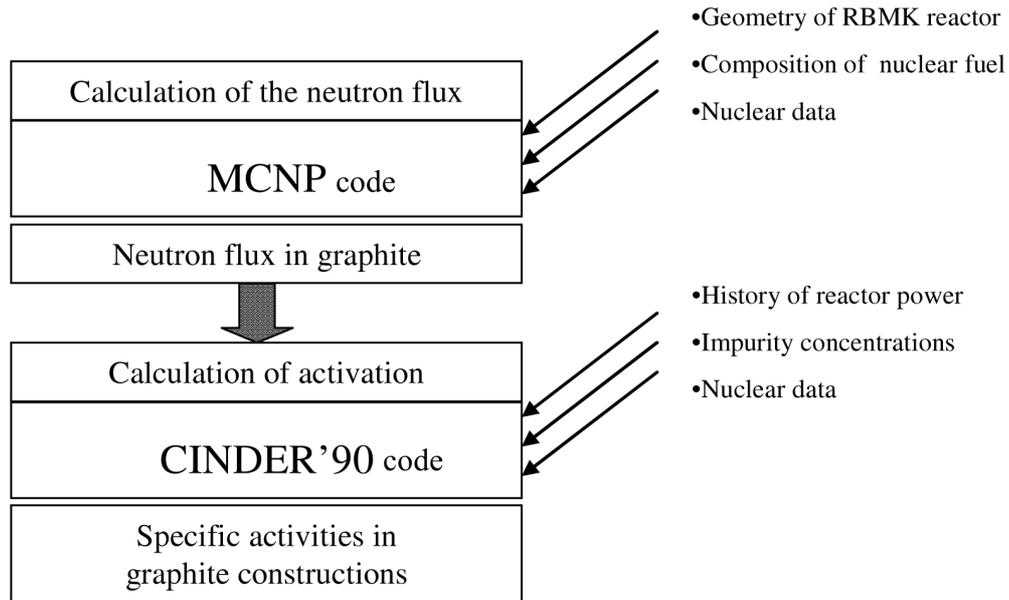


Fig. 1. The scheme for the calculations of the neutron flux and the activation of RBMK reactor graphite.

Table 1. Main radionuclides in the graphite of the Ignalina NPP reactor (21 years of its operation and one year after the shutdown of Unit 1).

Impurity	Concentration in graphite, ppm	Reaction and the resulting radionuclide	Averaged specific activity, Bq·g <sup>-1</sup>	
			Moderator	Reflector
<sup>6</sup> Li	0.002	<sup>6</sup> Li(n,α)→ <sup>3</sup> H	2.5·10 <sup>5</sup>	2.4·10 <sup>5</sup>
<sup>13</sup> C	11100	<sup>13</sup> C(n,γ)→ <sup>14</sup> C	5.1·10 <sup>5</sup>	1.2·10 <sup>5</sup>
<sup>14</sup> N	35	<sup>14</sup> N(n,p)→ <sup>14</sup> C		
<sup>35</sup> Cl	5.76	<sup>35</sup> Cl(n,γ)→ <sup>36</sup> Cl	3.9·10 <sup>3</sup>	1.4·10 <sup>3</sup>
<sup>54</sup> Fe	1.09	<sup>54</sup> Fe(n,γ)→ <sup>55</sup> Fe	5.8·10 <sup>5</sup>	1.5·10 <sup>5</sup>
<sup>59</sup> Co	0.019	<sup>59</sup> Co(n,γ)→ <sup>60</sup> Co	1.2·10 <sup>5</sup>	3.2·10 <sup>5</sup>
<sup>133</sup> Cs	0.0016	<sup>133</sup> Cs(n,γ)→ <sup>134</sup> Cs	2.8·10 <sup>3</sup>	3.2·10 <sup>3</sup>
<sup>153</sup> Eu	0.0014	<sup>153</sup> Eu(n,γ)→ <sup>154</sup> Eu	2.8·10 <sup>2</sup>	2.0·10 <sup>3</sup>
<sup>154</sup> Eu*	–	<sup>154</sup> Eu(n,γ)→ <sup>155</sup> Eu	2.1·10 <sup>2</sup>	1.3·10 <sup>3</sup>

\* formed as the result of the reaction <sup>153</sup>Eu(n,γ)→<sup>154</sup>Eu

carbon cycle: the atmosphere, the biosphere, the hydrosphere, and the lithosphere. In order to evaluate the radiological consequences we considered a critical group of people residing in the area at a distance of 1 km from the incineration facility (border of assumed sanitary protection zone) and spending all the time in the small sector (width 22.5°) in the prevailing wind direction.

Radionuclides in the plume at the location of interest would cause an external exposure dose to the population due to cloud immersion and internal exposure due to its inhalation. Radiocarbon will be assimilated from the air by photosynthesis of the local flora; some fraction of it will be deposited on the ground due to washout from the atmosphere by rain. It will enter the food chain and will cause an internal exposure due to ingestion of contaminated food products.

Transport of the released plume (smoke) is modelled

using a Gaussian dispersion (plume) model. Volumetric activity of the radionuclide in air  $C_{\text{air}}(x, y, z, h_e)$  is evaluated for the downwind direction centre line of the cloud at the distance  $x$  ( $x$  axis direction,  $y = 0$ ) at the height of interest  $z$ , i. e., 1 m above ground ( $z = 1$  m). The initial elevation of the release source is taken into account:

$$C_{\text{air}}(x, y, z, h_e) = \frac{\dot{Q}}{2\pi \cdot \sigma_y \sigma_z \cdot \bar{u}} V(z, h_e, L, \sigma_z),$$

$$V(z, h_e, L, \sigma_z) = \sum_{n=-5}^5 \left\{ \exp \left[ -\frac{1}{2} \left( \frac{z - h_e + 2nL}{\sigma_z} \right)^2 \right] + \exp \left[ -\frac{1}{2} \left( \frac{z + h_e + 2nL}{\sigma_z} \right)^2 \right] \right\}, \tag{1}$$

where  $\dot{Q}$  is the radiocarbon release rate from the

incinerator stack,  $\text{Bq}\cdot\text{y}^{-1}$ ;  $\sigma_y$ ,  $\sigma_z$  are the standard deviations of the horizontal and vertical Gaussian distribution, or dispersion coefficients, m;  $\bar{u}$  is the average wind speed during the time of release,  $\text{m}\cdot\text{y}^{-1}$ ;  $h_e$  is the effective release height, m;  $L$  is the height of boundary layer, m;  $n$  is a summation index. The activity derived from Eq. (1) applies to the dispersion of material, which is not removed from the plume as it travels downwind (conservative assumption). Horizontal and vertical standard deviations  $\sigma_y$  and  $\sigma_z$  depend on the distance from the source ( $x$  is given in metres), atmosphere stability (Pasquill) class  $s$ , and are calculated by the following equations:

$$\begin{aligned}\sigma_y(x, s) &= p_y(s) \cdot (x)^{q_y(s)}, \\ \sigma_z(x, s) &= p_z(s) \cdot (x)^{q_z(s)},\end{aligned}\quad (2)$$

where  $p_y(s)$ ,  $q_y(s)$ ,  $p_z(s)$ , and  $q_z(s)$  are the coefficients for six different atmosphere stability classes (from A to F). The coefficients are presented in Table 2.

Basic scenario of the graphite incineration assumes that it is continuously burnt during one year and all inventory of  $^{14}\text{C}$  is released into the atmosphere in the form of  $\text{CO}_2$ . For the estimation of the external exposure dose from cloud and the inhalation dose it is essential to consider frequency of the wind blowing towards the sector of interest and occurrence of the atmosphere stability class during the year. The most prevalent is southwest wind in Lithuania. The wind is blowing from the facility on the average  $P_a = 13.3\%$  of the time during the year [6]. Typical frequency of the certain atmosphere stability class occurrence  $n_s$  during the year observed in Lithuania is used in the calculations (see Table 2).

An average annual external exposure dose from the plume (cloud submersion) and the inhalation dose at the location of  $x = 1000$  m from the source (assuming continuous releases during the year; initial elevation of the plume  $h_e = 20$  m and the resulting concentrations are distributed evenly across a  $\Delta\Theta = 22.5^\circ$  sector) are calculated by the following equations:

$$\begin{aligned}D_{\text{ext\_cloud}} &= DFact_{\text{ext\_cloud}} \cdot P_a \cdot \bar{C}_{\text{air}}, \\ D_{\text{inh}} &= DFact_{\text{inh}} \cdot R_{\text{inh}} \cdot P_a \cdot \bar{C}_{\text{air}},\end{aligned}\quad (3)$$

where

$$\bar{C}_{\text{air}} = \frac{\dot{Q} \cdot \Delta t}{2\pi} \sum_{s=1}^6 \frac{n_s}{\sigma_y(x, s) \cdot \sigma_z(x, s) \cdot \bar{u}(s)}$$

is the integral volumetric air activity at the place of interest,  $\text{Bq}\cdot\text{y}\cdot\text{m}^{-3}$ ;  $DFact_{\text{ext\_cloud}} = 9.36 \cdot 10^{-15}$  ( $\text{Sv}\cdot\text{h}^{-1}$ )/( $\text{Bq}\cdot\text{m}^{-3}$ ) and  $DFact_{\text{inh}} = 2.4 \cdot 10^{-3}$   $\text{Sv}\cdot\text{Bq}^{-1}$

[7] are dose factors for  $^{14}\text{C}$  radionuclide cloud submersion and inhalation, respectively;  $R_{\text{inh}} = 0.93 \text{ m}^3\cdot\text{h}^{-1}$  is the average inhalation rate by an adult [7];  $\Delta t$  is duration of the release (1 year).

The estimated inhalation dose for an adult permanently residing at the location within one sector at the distance  $x = 1000$  m is equal to  $5.2 \cdot 10^{-6}$  Sv. The external exposure dose from the cloud submersion is negligible ( $2.7 \cdot 10^{-11}$  Sv).

Part of  $^{14}\text{CO}_2$  will be assimilated by the plants due to photosynthesis if the release occurs continuously during the year. For the continuous releases, transfer of  $^{14}\text{C}$  is evaluated using the specific activity methodology, which assumes that the ratio of  $^{14}\text{C}$  in food to the ratio of  $^{14}\text{C}$  in air is the same as the ratio of stable carbon isotopes in air. Transfer factors for radiocarbon transfer from the air to the main food products are given in Table 3. The effective dose of the adult member of the critical group in the location is calculated assuming that all food products were produced locally. The ingestion dose is calculated by the following equation:

$$D_{\text{ing}} = DFact_{\text{ing}} \cdot \bar{C}_{\text{air}} \cdot \sum_m TF_m \cdot q_m, \quad (4)$$

where  $D_{\text{ing}}$  is the effective dose due to food ingestion, Sv;  $DFact_{\text{ing}} = 5.8 \cdot 10^{-10}$   $\text{Sv}\cdot\text{Bq}^{-1}$  is the ingestion dose factor for  $^{14}\text{C}$  [7];  $m$  is the index for the food product type;  $TF_m$  is the transfer factor of  $^{14}\text{C}$  from air to  $m$ th type food product,  $\text{m}^3\cdot\text{kg}^{-1}$ ;  $q_m$  is the  $m$ th type food product consumption rate,  $\text{kg}\cdot\text{y}^{-1}$  (see Table 3). The estimated effective dose due to ingestion of  $^{14}\text{C}$  with contaminated foods for the basic release scenario is 2.7 mSv.

If the radioactive release is occurring in wintertime when vegetation is not growing, there will be no direct assimilation of  $\text{CO}_2$  from the atmosphere by the plants. In this case  $^{14}\text{C}$  can enter food chain only if  $^{14}\text{CO}_2$  will be washed out from the atmosphere by the rain and would be deposited on soil. Part of the deposited  $^{14}\text{C}$  will be assimilated by the plants during next vegetation period.  $\text{CO}_2$  is little soluble in water under normal conditions. Fraction of  $\text{CO}_2$  which is washed out from the atmosphere can be predicted by the simple upper-limit estimate, assuming that the raindrops are in equilibrium with the air phase when they reach ground. This assumption is supported by analysis of Cohen [9] who has shown that raindrops having fallen through 10 meters of the polluted atmosphere will be nearly at equilibrium with the gaseous phase (provided that the contaminant is not reactive in the aqueous rain phase). It is essentially true for  $\text{CO}_2$  having in mind that the ratio of diluted  $\text{CO}_2$  to  $\text{H}_2\text{CO}_3$  is 400:1 in the water [10].

Table 2. Atmosphere parameters for various stability classes  $s$ : the typical frequency of the certain atmosphere stability class occurrence  $n_s$  observed in Lithuania during the year; the height of boundary layer  $L$ ; an average wind speed  $\bar{u}(s)$ ; coefficients for the plume dispersion calculations at a height of  $h_e = 20$  m above ground.

Stability class $s$	Fraction of time of the certain stability class occurrence $n_s$ , % [6]	Boundary layer $L(s)$ , m [6]	Wind speed $\bar{u}(s)$ , $\text{m}\cdot\text{s}^{-1}$ [6]	Coefficients			
				$p_y(s)$	$q_y(s)$	$p_z(s)$	$q_z(s)$
A	0.72	1600	4.3	1.503	0.833	0.151	1.219
B	6.77	1200	4.7	0.876	0.823	0.127	1.108
C	10.2	800	5.6	0.659	0.807	0.165	0.996
D	56.7	560	12.1	0.640	0.784	0.215	0.885
E	8.49	320	10.9	0.801	0.754	0.264	0.774
F	16.9	200	4.9	1.294	0.718	0.241	0.662

Table 3. Transfer factors  $TF_m$  for continuous releases of  $^{14}\text{C}$  derived using the specific activity method  $C_{^{14}\text{C}}(\text{food}) = C_{^{14}\text{C}}(\text{air}) \cdot TF$ , and consumption rates per person of the food products.

[-] Food products	Transfer factor, $\text{m}^3\cdot\text{kg}^{-1}$ , [4]	Consumption rate, $\text{kg}\cdot\text{y}^{-1}$ , [8]
Beef	1188	29
Cereal	2225	124
Eggs	938	12.7
Fruit (domestic and wild)	241	61
Green vegetables	231	45
Leguminous vegetables	556	50
Milk	294	287
Pork	1638	30
Potato	563	118
Poultry	844	20
Root vegetables	194	26

Thus, the solubility of the  $\text{CO}_2$  gas in the rainwater can be estimated by the Henry's law:

$$M_{\text{CO}_2\text{-rain}} = K_{\text{H}}(T) \cdot p_{\text{CO}_2}, \quad (5)$$

where  $M_{\text{CO}_2\text{-rain}}$  is the molar concentration of the dissolved  $\text{CO}_2$  in the rainwater,  $\text{mol}\cdot\text{l}^{-1}$ ;  $K_{\text{H}}(T = 273 \text{ K}) = 0.077 \text{ mol}\cdot\text{l}^{-1}\text{atm}^{-1}$  is the temperature dependent Henry's constant;  $p_{\text{CO}_2} = 0.000367 \text{ atm}$  is the  $\text{CO}_2$  partial pressure in the atmosphere under normal conditions. Some  $\text{CO}_2$  molecules will be formed by  $^{14}\text{C}$  isotope. The estimated average  $^{14}\text{C}$  activity annually dissolved in the rainwater is calculated by the following equation:

$$C_{^{14}\text{C}\text{-rain}} = M_{\text{CO}_2\text{-rain}} \cdot K \cdot \frac{\bar{C}_{\text{air}}}{\eta_{\text{CO}_2}}, \quad (6)$$

where  $C_{^{14}\text{C}\text{-rain}}$  is the  $^{14}\text{C}$  volumetric activity in the rainwater at the location of interest,  $\text{Bq}\cdot\text{y}\cdot\text{l}^{-1}$ ;  $K = 1/0.0224 \text{ m}^3\cdot\text{mol}^{-1}$  is the conversion coefficient estimated for gas under normal conditions;  $\eta_{\text{CO}_2}$  is the volumetric part of  $\text{CO}_2$  in the air. Total wet deposition of  $^{14}\text{C}$  on soil is evaluated by the following relationship:

$$D_{^{14}\text{C}} = h \cdot C_{^{14}\text{C}\text{-rain}}, \quad (7)$$

where  $D_{^{14}\text{C}}$  is the radiocarbon activity in the depositions,  $\text{Bq}\cdot\text{m}^{-2}$ ;  $h$  is the annual rate of rain at the location,  $\text{mm}\cdot\text{y}^{-1}$ . It is assumed that the annual rate of rain is  $h = 500 \text{ mm}\cdot\text{y}^{-1}$  (average annual amount of precipitation in Lithuania is 662 mm) and all the events of rain have occurred when wind has been blowing from the incinerator (conservative assumption). The estimated deposition density of  $^{14}\text{C}$  for the basic scenario at the locations  $x = 1000 \text{ m}$  is of the order of  $49.5 \text{ Bq}\cdot\text{m}^{-2}$ .  $^{14}\text{C}$  deposited on soil will migrate to deeper soil layers, part of it will be assimilated by the plants growing in the area, and the remainder will be released back to the atmosphere by the respiration. The dose due to  $^{14}\text{C}$  depositions is assessed using a dynamic RESRAD 6.22 model [11]. It takes into account the main processes of the radiocarbon behaviour in the soil for the Lithuanian conditions. The calculated dose includes the external exposure dose from ground, the dose due to inhalation of the released radiocarbon back to the atmosphere, and the dose due to ingestion of the food products originated from the contaminated soil. Consumption rates of the food products are presented in Table 3. Calculations show that in the first year after the deposition the dose is  $0.88 \mu\text{Sv}$ . The value decreases by about two orders of magnitude for the subsequent year.

## 5. Global scale radiological consequences

As  $^{14}\text{C}$  has a long half-life, the fluxes of radiocarbon and stable carbon among different reservoirs (the atmosphere, the biosphere, the hydrosphere, and the lithosphere) are governed by the same exchange processes. A number of models have been developed to study global circulation of stable carbon, especially due to recent increased interest in effects of the climate change. Some of them can be directly used for the assessment of the radiological consequences of the naturally produced and man-made sources of  $^{14}\text{C}$ . Compartment dynamic models are sufficiently accurate for such purposes of the long-term collective dose commitment assessment. The models predict time dependent activities per gram of carbon in each environmental compartment. It is assumed that the specific activity of  $^{14}\text{C}$  isotope in carbon ingested by humans is the same as that in the most relevant compartments for food intake (ground vegetation for terrestrial foods and relevant surface ocean compartments for marine foods). Ingestion of  $^{14}\text{C}$  with the food contributes to 99% of the total dose caused by this radionuclide.

A model developed by Titley et al. [12] is widely recognized as the most relevant for the global  $^{14}\text{C}$  modelling [13]. It contains 23 compartments (Fig. 2): atmosphere, ocean sediments, the Antarctic ocean (four layers), the Atlantic Ocean (four layers), the Pacific Ocean, including the Indian ocean (three layers), the Arctic Ocean (two layers), woody tree parts, non-woody tree parts, ground vegetation, decomposers, soil, and a compartment representing input from fossil fuel (gas, coal, oil) burning.

Exchange between the atmosphere and the terrestrial biosphere is based on estimates of the global photosynthesis uptake of carbon by plants and its release to the atmosphere by respiration of plants, animals, and from soil [14, 15]. The carbon behaviour in the ocean is modelled by taking into account temperature changes, surface area, and varying area of ice cover in winter. Photosynthesis at the surface of the ocean and subsequent transfer of carbon down the water column are significant effects and are also taken into account in the model. Evaluations of the average annual doses per person and the cumulative dose for the global population due to release of the total radiocarbon inventory into the atmosphere were performed using the model. The obtained doses are presented in Table 4.

The incineration of all the graphite results in an additional global dose of  $0.0053 \mu\text{Sv}$  per person for the first year after release, and the cumulative dose of  $0.43 \mu\text{Sv}$

Table 4. Doses for the world population from the releases of  $7 \cdot 10^{14}$  Bq of  $^{14}\text{C}$  into the atmosphere from the incinerator.

Year	Average effective dose per person, $\mu\text{Sv}$	
	Annual dose	Cumulative dose
1	$5.3 \cdot 10^{-3}$	$5.3 \cdot 10^{-3}$
2	$1.7 \cdot 10^{-2}$	$2.3 \cdot 10^{-2}$
5	$2.7 \cdot 10^{-2}$	$9.1 \cdot 10^{-2}$
10	$1.7 \cdot 10^{-2}$	$1.9 \cdot 10^{-1}$
20	$7.7 \cdot 10^{-3}$	$2.9 \cdot 10^{-1}$
50	$3.9 \cdot 10^{-3}$	$4.3 \cdot 10^{-1}$
100	$2.5 \cdot 10^{-3}$	$5.9 \cdot 10^{-1}$
200	$1.6 \cdot 10^{-3}$	$7.7 \cdot 10^{-1}$
500	$9.8 \cdot 10^{-4}$	1.2
1000	$7.7 \cdot 10^{-4}$	1.5
2000	$6.1 \cdot 10^{-4}$	2.1
5000	$4.1 \cdot 10^{-4}$	3.4
10000	$2.2 \cdot 10^{-4}$	4.9
20000	$6.2 \cdot 10^{-5}$	5.9
50000	$1.4 \cdot 10^{-6}$	6.5

per person for the period of 50 years. The model predicts the collective dose committed in 50000 years to be equal to 65000 man·Sv, assuming that the future world population stabilizes at  $10^{10}$  people in the middle of this century. However, this value can vary in the range of 56000–92000 man·Sv if one takes into account uncertainty of the man activities (e. g., burning of fossil fuels) and climate changes [13].

## 6. Selection of the optimal scenario for incineration

Summary of the estimated effective doses per unit activity release (1 Bq) and due to incineration of 1700 tons of used graphite in the reactor of the INPP Unit 1 is presented in Table 5. All the dose estimates are derived using conservative assumptions of low stack height, place of residence, and local origin of all food products.

The local population will receive the largest effective dose fraction due to consumption of the locally produced food products, namely 2.7 mSv. The value is in a relatively good agreement with the one of 0.2 mSv evaluated by other authors [3] for releases under similar conditions of  $6 \cdot 10^{14}$  Bq of  $^{14}\text{C}$ . The value exceeds a limit of  $0.2 \text{ mSv} \cdot \text{y}^{-1}$  set by the national radiation safety normative document [16] for the projected nuclear installations. Continuous releases of  $^{14}\text{C}$  from the incinerator would be acceptable only if graphite from Unit 1 were incinerated over the extended period of time (not shorter than 14 years). The fraction of the dose due to ingestion could be eliminated if the incinerator operated in the dark time of the day or in wintertime when local vegetation does not perform photosynthesis. It

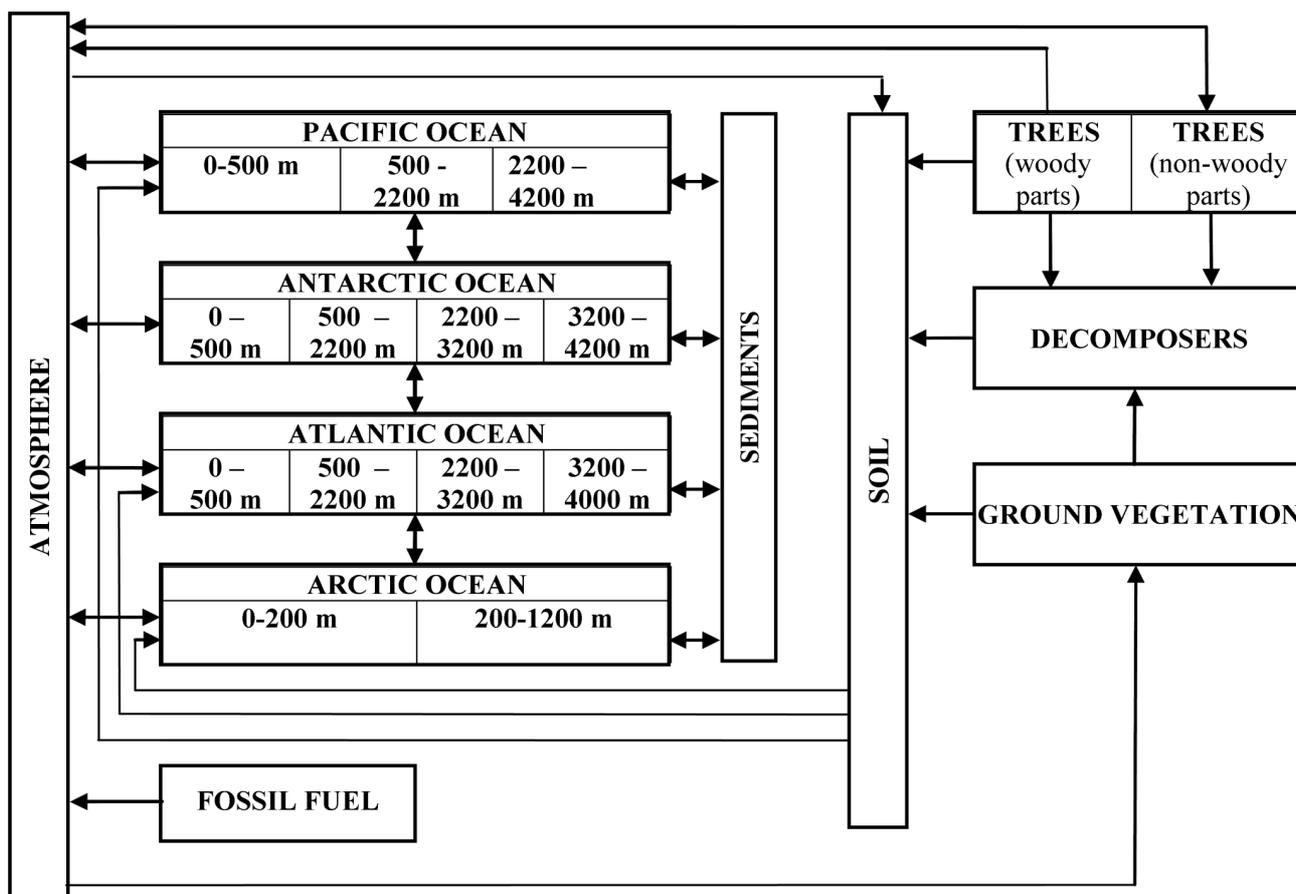


Fig. 2. Compartment model for global circulation of  $^{14}\text{C}$  [12] used in the calculations.

would also be reasonable to operate the facility in the absence of rain in order to eliminate fraction of the dose due to washout of  $^{14}\text{CO}_2$  from the plume and its deposition on the soil. The optimal scenario would be incineration of graphite in winter with no rain. It would cause the effective dose only of  $5.2 \mu\text{Sv}$  for the local population.

The radiological impact on the world population is very small.  $^{14}\text{C}$  is a radionuclide naturally produced in the stratosphere and it is globally dispersed all over the world. The natural production rate of radiocarbon is  $1 \cdot 10^{15} \text{ Bq} \cdot \text{y}^{-1}$  by the action of the cosmic neutrons in the atmosphere. Inventory of  $^{14}\text{C}$  in the atmosphere is about  $1.2 \cdot 10^{17} \text{ Bq}$ . It leads to an average individual dose rate of  $12 \mu\text{Sv} \cdot \text{y}^{-1}$  [13]. Release of  $7 \cdot 10^{14} \text{ Bq}$  from the incinerator is less than the annual natural production of radiocarbon and it is 0.6% of its total inventory in the atmosphere. The annual dose for the world population due to the total graphite inventory incineration is small. The maximum increase (during the fifth year after the release) of world population exposure is only 0.001% of the natural background.

Despite estimates that operation of the used graphite

incinerators has only a minor radiological impact on the population, such facilities are not operating anywhere in the world. It is because of the prevailing negative opinion of the public to have such facilities in the neighbourhood.

## 7. Conclusions

The problem of nearly 1700 tons of irradiated graphite, originating from dismantling of the Ignalina NPP RBMK-1500 reactor, was discussed in this work. The radiological consequences of  $^{14}\text{C}$  due to graphite incineration were evaluated. The estimated maximal inventory of the radionuclide  $^{14}\text{C}$  in the reactor of the Ignalina NPP Unit 1 is  $\sim 7 \cdot 10^{14} \text{ Bq}$ .

Continuous incineration of 1700 tons of irradiated graphite from the Ignalina NPP RBMK-1500 reactor in the especially designed incinerator would cause an additional average effective dose of 2.7 mSv for the critical group of local population. In order not to exceed limits of radiation safety norms set by legislation in Lithuania, incineration should be done in a period not shorter than 14 years. However, the radiological

Table 5. Contributions of different pathways to effective doses for local population due to continuous releases of  $^{14}\text{C}$  from the incinerator during one year.

Exposure pathway	Dose per unit release (1 Bq) of $^{14}\text{C}$ from the incinerator, Sv·Bq $^{-1}$	Dose due to release of total inventory ( $7\cdot 10^{14}$ Bq) of $^{14}\text{C}$ from the incinerator, Sv
Ingestion of foodstuffs	$3.84\cdot 10^{-18}$	$2.7\cdot 10^{-3}$
Inhalation	$7.40\cdot 10^{-21}$	$5.2\cdot 10^{-6}$
Wet deposition	$1.25\cdot 10^{-21}$	$8.8\cdot 10^{-7}$
Plume shine	$5.26\cdot 10^{-25}$	$3.7\cdot 10^{-11}$
Total	$3.84\cdot 10^{-18}$	$2.7\cdot 10^{-3}$

impact could be significantly reduced if the incinerator operated only in the dark time of the day or in winter. This would be the optimal scenario for the radioactive graphite incineration, which would cause the additional dose only of  $5.2\ \mu\text{Sv}$  for the local population. World population would receive an average lifetime (50 years) dose of  $0.43\ \mu\text{Sv}$  per person which is negligibly small.

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## $^{14}\text{C}$ IZOTOPU UŽTERŠTO IGNALINOS AE GRAFITO DEGINIMO RADIACINIŲ PASEKMIŲ VERTINIMAS

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### Santrauka

Nutraukus Ignalinos AE pirmojo bloko reaktoriaus eksploataciją, aktualia tapo radioaktyviųjų atliekų šalinimo problema. Be kitų radioaktyviųjų atliekų, kiekviename reaktoriuje yra apie 1700 tonų grafito, kuris užterštas  $^{14}\text{C}$  radioizotopu. Maksimalus  $^{14}\text{C}$  radionuklido įvertintas aktyvumas pirmojo bloko reaktoriaus grafite yra maždaug  $7 \cdot 10^{14}$  Bq. Vienas iš galimų šio grafito utilizacijos būdų yra jo sudeginimas specialiaame įrenginyje. Tokiu atveju į atmosferą būtų išmesti nemaži  $^{14}\text{C}$  radionuklido kiekiai  $\text{CO}_2$  dujose. Į aplinką patekusi radioaktyvioji anglis sukeltų papildomą gyventojų apšvitą. Galimos radiacinės pasekmės Lietuvos gyventojams įvertintos modeliuojant radioaktyviosios anglies sklaidą aplinkoje įvairiems grafito deginimo scenarijams. Deginimo metu susidariusi dujų sklaida atmosferoje modeliuojama taikant Gauso modelį. Įvertinama  $\text{CO}_2$  asimiliacija augalams vykdant fotosintezę,  $\text{CO}_2$  iš-

plovimas iš atmosferos lietumi, iškritusios  $^{14}\text{C}$  pereiga iš dirvožemio į augmeniją ir maisto produktų tarša. Nustatyta, kad nepertraukiamai deginant grafitą specialiaame įrenginyje, kritinė vietinių gyventojų grupė (gyvenančių už 1 km nuo šaltinio) papildomai gautų apie 2,7 mSv efektingą apšvitą dozė. Didžiausią dozės dalį lemia užterštų vietinių maisto produktų vartojimas. Toks grafito utilizacijos būdas būtų priimtinas (neviršytų  $0,2 \text{ mSv} \cdot \text{metai}^{-1}$ ), jeigu visas pirmojo bloko reaktoriaus grafitas būtų sudegintas ne trumpiau kaip per 14 metų. Mažiausias radiacines pasekmes gyventojams turėtų reaktoriaus grafito deginimas žiemą arba tamsiu paros metu (kai nevyksta fotosintezė). Toks grafito sudeginimo scenarijus lemtų  $5,2 \mu\text{Sv}$  vidutinę efektingą dozę kritinei vietinių gyventojų grupei. Papildoma vidutinė apšvitą dozė planetos gyventojui neviršytų  $0,43 \mu\text{Sv}$  per gyvenimą (50 metų).