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APPRAISAL METHOD FOR THE POPULATION RADIATION DOSE FROM ^{14}C EMISSIONS OF NUCLEAR ENTERPRISES

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The yearly effective population dose of radiation in Rezhik Village on account of ^{14}C emissions from nuclear power facilities situated in the immediate vicinity is appraised. The method of appraisal is based on an analysis of the specific activity of ^{14}C in tree rings in the vicinity of the enterprises. The impact of the ^{14}C radiation on the population was appraised with and without radiocarbon balance between atmospheric air, food, and the human body. The range of the annual effective radiation dose to the population dose was equal to 0.89–2.23 μSv in the state of ^{14}C equilibrium and 0.08–0.33 μSv away from equilibrium. The technogenic contribution to the annual effective radiation dose from ^{14}C was equal to 4.4–15.5%.

^{14}C is a low-energy β-emitting radionuclide produced both naturally and technogenically. The natural route of formation is based on the interaction of thermal neutrons and ^{14}N atoms in the upper layers of the tropo- and stratospheres. The annual production of ^{14}C via the reaction $^{14}N(n, p)$ is equal to 1.4 PBq.^{14}C The total amount of ^{14}C in the atmosphere is estimated to be 140 PBq [1]. The technogenic input of radiocarbon in the 1950s–1960s, estimated to be 220 PBq, was due to atmospheric tests of nuclear weapons. After ground-based nuclear explosions were banned, the main anthropogenic source of ^{14}C in the environment became emissions from nuclear power facilities. Regular monitoring of radiocarbon emissions in our country has been conducted only for the last seven years. There are no data before 2015, or information on emissions is presented only for the period of research [2]. In connection with this, it is now important to determine ^{14}C releases as well as the population dose rate resulting from the operation of nuclear reactors before the start of regular mandatory monitoring of ^{14}C in the emissions.

It is shown in [3] that ^{14}C is one of the main dose-forming radionuclides in NPP emissions. In an operating nuclear reactor ^{14}C is formed mainly on the interaction of thermal neutrons with ^{14}N, ^{13}C, ^{17}O atoms present in structural materials, moderator, and coolant as well as on uranium and plutonium ternary fission reactions in the fuel. As a biogenic analogue of stable ^{12}C, radiocarbon penetrates into the organs and tissues of living organisms and is incorporated into the composition of fats, carbohydrates, protein molecules, and genetic structures. The danger of its incorporation comes from the internal irradiation of molecules of organic compounds and mutagenic disorders due to the conversion of ^{14}C into^{14}N, which underscores the importance of controlling dose loads [4]. In 2015, ^{14}C was placed on the list of pollutants subject to state regulations pertaining to environmental protection.

Comprehensive investigations, performed over many-years, of stationary sources of emissions of radioactive substances from nuclear power plants have demonstrated the need for constant monitoring of the release of ^{14}C from ventilation systems into the atmosphere [5]. Continuous monitoring of the source of release ensures compliance with the standards for maximum allowable releases of radioactive substances, guaranteeing compliance with population dose quotas. The absence of results obtained by regular monitoring of ^{14}C in NPP emissions precludes direct assessment of the radiation impact for the

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TABLE 1. Specific Activity of ^{14}C in Annual Rings of Pines Growing in the Area of the Beloyarsk NPP and the Institute of Reactor Materials (IRM), Bq/kg C [6]

Year	Sampling location	Background	ΔA
1970	364.1 ± 10.9	336.7 ± 1.3	27.4 ± 0.8
1980	343.6 ± 34.5	278.0 ± 6.3	65.6 ± 6.8
1990	297.3 ± 24.6	260.7 ± 2.2	36.6 ± 3.0
1993	281 ± 0.3	250.5 ± 0.2	30.5 ± 0.1
1994	330.3 ± 7.4	260.3 ± 1.6	70 ± 1.3
1995	392.6 ± 48.6	263.9 ± 0.8	128.7 ± 11.3
2000	340.2 ± 6.9	242.1 ± 0.6	98.1 ± 1.7
2005	346.2 ± 12.8	234.3 ± 2.3	111.9 ± 3.1
2010	267.9 ± 8	236.7 ± 2.1	31.2 ± 0.7

years of NPP operation up to 2018. Of particular interest is the possibility of calculating the effective population dose in the absence of constant monitoring of ^{14}C in emissions from nuclear energy facilities.

This article presents estimates of the radiation effect of ^{14}C emissions on the population of Rezhik Village (Sverdlovsk Oblast) in 1970–2010 as a result of the operation of the AMB-100 and -200 uranium-graphite reactor units, a BN-600 fast reactor at the Beloyarsk NPP, and the IVV-2M reactor at the Institute of Reactor Materials in the Zarechnyi Town (Sverdlovsk Oblast). The estimate was made by analyzing the specific activity of ^{14}C in the annual rings of pine trees growing in the area of the emission sources [6].

Materials and procedures. Estimation of ^{14}C release involves the sequential solution of the following problems:

– selection of the source of entry into the environment; in this article, the nuclear reactor AMB-100, -200, BN-600, and IVV-2M, situated on the same industrial site, are selected as sources. AMB-100 was operated in 1964–1981 and AMB-200 in 1969–1989; IVV-2M startup took place in 1966, but it began to be taken into account in1995 as a comparable source of ^{14}C – the start of the commercial production of radiocarbon;

– determination of the area with the greatest accumulation in woody vegetation;

– calculation of the dilution factor, which determines the ratio of the average annual volumetric activity in the surface air layer to the average annual emission rate into the atmosphere according to the recommendations [7];

– sampling of annual rings in a selected area using a Pressler augur and measuring ^{14}C activity in annual rings;

– analysis of the results according to the calculation procedure;

– calculation of the dilution factor, ^{14}C emission from the site and the annual effective population dose in Rezhik Village.

The model of ^{14}C transport into the environment based on the principle of isotopic equilibrium was used in the calculations, i.e., the specific activity of radiocarbon in plants corresponds to the specific activity in the ambient air [8]. This approach has been implemented in studies of the specific activity of atmospheric radiocarbon based on the specific activity of ^{14}C in plants, including annual rings, which are in equilibrium with the environment [9–13].

The specific activity of ^{14}C in the annual rings of 10 kinds of >60 yr old pines growing in the area under consideration is given in [6] (Table 1). Rings of same year of all trees were combined into a single sample, divided into two samples, and the ^{14}C activity was measured at the Center for Collective Use "Cenozoic Geochronology" of the Institute of Archeology and Ethnography of the Siberian Branch of the Russian Academy of Sciences, using the acceleration mass spectrometer at the Institute of Nuclear Physics of the Siberian Branch of the Russian Academy of Sciences. The specific activity of ^{14}C in the annual rings of a 113-year-old pine grown in Akademgorodok (Novosibirsk) was used as the background activity [14].

The Rezhik population is chosen as a critical group based on the following factors: rural type (largest use of local products), situated in line with the sampling site and the most repeatable wind direction.

Computational procedure. The average annual volumetric activity of ^{14}C in the atmospheric air at the critical site of the emission sources was calculated by the formula

Fig. 1. Arrangement of ^{14}C emission sources and sampling site [6].

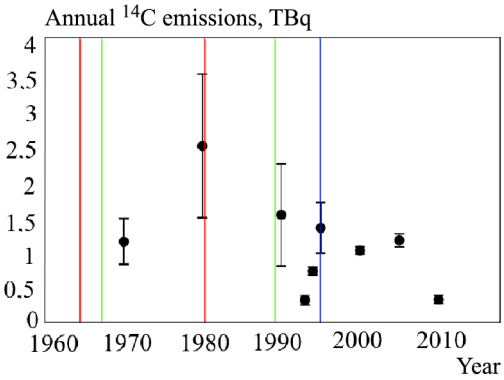

Fig. 2. Estimated annual emissions of ^{14}C: AMB-100 in 1964–1981(—), AMB-200 in 1967–1989 (—), ^{14}C production at IRM since 1995 (—) (the bounds show the confidence interval).

$$A_V = \Delta A C_{st},$$

where ΔA is the difference between measured and background specific activity, Bq/kg C; C_{st} is the concentration of stable carbon in the air, kg/m^3. The concentration of stable carbon is equal to 200 mg/m^3 according to [15] and 160 mg/m^3 according to [16]. In the present article, the concentration was taken as 180 mg/m^3.

The annual ^{14}C emission was estimated by the formula

$$Q = A_V 3.15 \cdot 10^7 / G_{n,i}, \quad (1)$$

where $3.15 \cdot 10^7$ is a conversion factor, sec/yr; $G_{n,i}$ is the average annual meteorological dilution factor in the surface layer of the atmosphere at distance x from the ith source in the direction of the wind at the nth rhumb, sec/m^3 (Fig. 1). In the calculations, AMB-100, -200 emissions of height 100 m and IRM 40 m were considered. For the sampling site, the dilution factor was $1.4 \cdot 10^{-7}$ for the Beloyarsk NPP and $5.5 \cdot 10^{-7}$ for IRM. For Rezhik village, values are $7.9 \cdot 10^{-8}$ and $1.7 \cdot 10^{-7}$ sec/m^3, respectively [17].

The currently used methods of calculating the maximum allowable emission are based on a conservative approach, wherein the diet consists only of local products so that the specific activity of ^{14}C in human tissues, food, and atmospheric air is observed to be in equilibrium. Then the expected annual internal radiation dose D from ^{14}C present as carbon dioxide $^{14}CO_2$ in the atmospheric air can be obtained using the formula [18]

$$D = A_V \mathrm{DCF} / C_{st}, \quad (2)$$

where DCF = $5.6 \cdot 10^{-5}$ (Sv/yr)/(Bq/g) is a conversion factor that correlates the annual internal radiation dose to the carbon dioxide concentration in human tissues per 1 g of stable carbon [19].

TABLE 2. Consumption of food products in the area of the Beloyarsk NPP

Food product	Annual consumption by the rural population, kg [18]	Relative consumption of local products, % [18]	Relative content of carbon in products, kg/kg [23]
Milk	269.5	8.5	0.065
Meat	79.8	22	0.2
Potato	51.1	92.1	0.046
Vegetables	103.7	69.5	0.059
Fruits	70.6	67.1	0.062

TABLE 3. Annual Effective Population Dose of 14C Radiation in Rezhik Village with a Conservative Approach (*1*) and Taking into Account the Actual Diet (*2*), μSv

Year	Background	Entry				
		Food		Inhalation, 10^{-4}	Total	
		1	*2*		*1*	*2*
1970	15.50 ± 0.09	0.89 ± 0.03	0.13 ± 0.01	1.43 ± 0.28	0.89 ± 0.03	0.13 ± 0.01
1980	13.05 ± 0.36	1.94 ± 0.2	0.29 ± 0.03	3.13 ± 0.89	1.94 ± 0.2	0.29 ± 0.03
1990	12 ± 0.11	1.18 ± 0.1	0.18 ± 0.01	1.91 ± 0.64	1.18 ± 0.1	0.18 ± 0.01
1993	11.53 ± 0.01	0.53 ± 0.01	0.08 ± 0.01	0.85 ± 0.01	0.53 ± 0.01	0.08 ± 0.01
1994	11.98 ± 0.08	1.21 ± 0.03	0.18 ± 0.01	1.96 ± 0.1	1.21 ± 0.03	0.18 ± 0.01
1995	12.14 ± 0.04	2.23 ± 0.28	0.33 ± 0.04	3.60 ± 0.68	2.23 ± 0.28	0.33 ± 0.04
2000	11.14 ± 0.03	1.7 ± 0.03	0.25 ± 0.01	2.74 ± 0.1	1.70 ± 0.03	0.25 ± 0.01
2005	10.78 ± 0.11	1.94 ± 0.07	0.29 ± 0.01	3.13 ± 0.18	1.94 ± 0.07	0.29 ± 0.01
2010	10.9 ± 0.1	0.54 ± 0.02	0.08 ± 0.01	0.87 ± 0.11	0.54 ± 0.02	0.08 ± 0.01

Since ^{14}C enters the human body mainly with food and the relative consumption of local products does not always exceed that of imported products, it is reasonable to assume that the specific activity of ^{14}C in human tissues and atmospheric air need at equilibrium. In this case, the expected annual radiation dose of ^{14}C can be calculated using the formula [20]

$$D = \varepsilon_{\text{inhal}} U A_V + \varepsilon_{\text{food}} \sum_i \alpha_i R_i A_{\text{food},i}, \quad (3)$$

where $\varepsilon_{\text{inhal}}$ is the dose conversion factor on inhalation of ^{14}C; for an adult, on inhalation of ^{14}C in the form of CO_2 it is equal to 6.2 pSv/Bq [21]; $U = 8.1 \cdot 10^3$ m^3/yr is the respiratory rate of an adult [22]; food $\varepsilon_{\text{food}} = 0.58$ nSv/Bq is the dose conversion factor for the dietary intake of ^{14}C for an adult [22]; α_i is the relative consumption of the *i*th local product in the population diet of the population (Table 2); R_i is the annual population consumption of the *i*th product, kg (see Table 2); $A_{\text{food},i}$ is the specific activity of ^{14}C in the *i*th local food product, Bq/kg.

The specific activity of ^{14}C in the *i*th food product, based on the balance of ^{14}C between atmospheric air and local plant or animal products, is calculated by the formula [23]

$$A_{\text{food},i} = A_V f_{p,i} / C_{\text{st}}, \quad (4)$$

where $f_{p,i}$ is the relative amount of carbon in the *i*th plant or animal product, kg/kg (see Table 2).

Results and discussion. The total annual intake of ^{14}C from various sources, as calculated by Eq. (1), is displayed in Fig. 2. The 1980 peak corresponds to the simultaneous operation of four power units with the reactors IVV-2M, AMB-100, -200, and BN-600. The annual emission activity decreased after the first power unit was decommissioned and then the second power unit of the first phase of the Beloyarsk NPP. A sharp increase in the annual ^{14}C emission in 1995 is associated with the

start of radiocarbon production at IRM. A subsequent reduction of the annual emission could be associated with technological improvements in the production of radioisotope products, followed by better handling of the products.

The population radiation dose loads from technogenic and native ^{14}C are given in Table. 3. The volumetric activity of native radiocarbon was taken from [14]. Calculations are made by two methods: under the condition that the diet consists only of local products – a conservative approach (see Eq. (2)) – and taking into account the actual diet (see Eqs. (3) and (4)). As can be seen from Table 3, the annual effective dose in the absence of equilibrium between atmospheric air and the human body is 6.7 times lower than with a conservative approach. Native radiocarbon makes the main contribution to irradiation.

The highest activity of the annual emissions of ^{14}C was observed in 1980 on four reactor units operating simultaneously – AMB-100, -200, BN-600, and IVV-2M – and after the first two units were shutdown in 1995 and IRM started commercial production of ^{14}C. The dose loads are minimal in the early 1990s, since they come from only two nuclear reactor units – IVV-2M and BN-600 – in regular operation. Today, the main source of ^{14}C in the area of the nuclear energy facilities considered in this article is the management of isotope products at IRM.

Conclusions. An assessment of the radiation impact of nuclear power plants makes it possible to reconstruct both the annual ^{14}C emission and its radiation impact on the population. The method employs accelerator mass spectrometry to analyze the specific activity of ^{14}C in the annual rings of trees standing in an area containing nuclear energy facilities.

The annual inflow of ^{14}C from the operation of nuclear power plants AMB-100, -200, and BN-600 amounted to 0.79–3.5 TBq. After the decommissioning of AMB-100 and -200, IRM became the main source of ^{14}C, equal to 0.26–2.2 TBq/yr. The annual ^{14}C emission in 2018 was equal to 0.34 TBq from IRM [24] and in 2020 1.02 GBq from BN-600 [25], which is consistent with the estimates. This indicates that currently the activities of IRM JSC are the main source of technogenic radiocarbon emissions into the environment.

The annual effective radiation dose to the population in Rezhik Village on account of ^{14}C emissions was equal to 0.53 ± 0.01 μSv in 1993 and 2.23 ± 0.28 μSv in 1995, based on a conservative approach wherein the diet consists only of local products. The approach based on the real diet made it possible to do away with conservatism, which reduced the dose loads by almost 7-fold: – to 0.08 ± 0.01 and 0.33 ± 0.04 μSv, respectively. The total annual dose from ^{14}C emissions does not exceed the minimum significant amount 10 μSv/yr in both approaches. The technogenic contribution to the annual effective population dose of ^{14}C radiation in Rezhik Village ranged from 4.4% in 1993 to 15.5% in 1995.

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