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Research Article

^{14}C in tree rings in the vicinity of the nuclear facility deployment areas*

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Abstract

^{14}C is naturally and artificially occurred radionuclide presented in atmosphere. ^{14}C is produced during the operation of a nuclear reactor of any type, enters the atmosphere and became a part of carbon cycle. The article presents the results of measuring the concentration of ^{14}C in the tree rings of 10 pines in the area of the Beloyarsk NPP (BelNPP) and the Institute of Nuclear Materials (INM), Zarechny. The sampling site, located 1200 m east of the INM, was selected based on long-term observations of meteorological parameters. The measurements were carried out using the accelerator mass spectrometer of the Budker Institute of Nuclear Physics, Novosibirsk. The influence of the operation of nuclear installations on the concentration of ^{14}C in the atmospheric air is demonstrated. The range of values for the concentration of carbon-14 in the sample ranged from 116.0 ± 4.4 to 192.0 ± 8.5 pMC.

Keywords

Radiocarbon, pine tree rings, accelerator mass spectrometer, nuclear reactor, nuclear power plant

Introduction

Currently, an important practical way to confirm the safe operation of nuclear power plants is the controlled release of radionuclides into the atmosphere. It is also necessary to monitor not only the source of radiation exposure, but also the environment affected (IAEA Safety Standards Series No. RS-G-1.8 2005). Each reactor plant type has a specific list of radionuclides that both define the total activity of the release and contribute mostly to the public exposure doses (Ekidin et al. 2016). One of the key radionuclides responsible for the public exposure dose formation is ^{14}C found in the release from nuclear reactors of any type (Nazarov et al. 2018). The Russian list of contaminants, to which state environmental regulation measures apply, includes ^{14}C (List of Pollutants). The IAEA treats ^{14}C as an

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important factor of the environmental and human radiation impacts in the process of the NPP operation (Management of Waste 2004, INPRO Methodology 2016).

In the 20th century, the key anthropogenic source of ^{14}C were nuclear weapon tests conducted in the period between 1945 and 1980. The total activity of ^{14}C that entered the atmosphere during the above period was about $3.5 \cdot 10^8$ GBq (Vasilenko et al. 1992).

At the present time, the major anthropogenic sources of ^{14}C are nuclear reactors and irradiated fuel reprocessing facilities. Operation of a nuclear reactor leads to ^{14}C forming largely as a result of the neutron activation reactions involving nuclei of various chemical elements contained in structural materials, fuel elements, moderator and coolant.

The major mechanisms for generation of ^{14}C in nuclear power reactors are (Management of Waste 2004)

a. the $^{14}N(n, p)^{14}C$ reaction with a very high thermal neutron capture cross-section of 1.82 barn (1 barn = $1 \cdot 10^{-24}$ cm^2);
b. the $^{17}O(n, \alpha)^{14}C$ reaction with a high thermal neutron capture cross-section (0.24 barn);
c. the $^{13}C(n, \gamma)^{14}C$ reaction with a small cross-section ($0.9 \cdot 10^{-3}$ barn);
d. ternary fission of fuel nuclei.

It has been counted that $1.1 \cdot 10^5$ GBq/y is released into the atmosphere as gases from all nuclear power plants in operation across the world, while about $3.7 \cdot 10^5$ GBq/y of ^{14}C is released in gaseous or liquid forms by spent nuclear fuel reprocessing facilities (Sources and Effects of Ionizing Radiation 2000).

Apart from ^{14}C entering the environment in a technological way, there is a natural path based on atoms of ^{14}N absorbing thermal neutrons which result from the interaction of cosmic rays with atmospheric substances: $^{14}N(n, p)^{14}C$. About $1.4 \cdot 10^6$ GBq of radiocarbon forms annually in such a way, and the total amount of ^{14}C in the atmosphere is estimated at $1.4 \cdot 10^8$ GBq. Most of ^{14}C is contained in oceans (about $1.0 \cdot 10^{10}$ GBq) (Management of Waste 2004).

The entry of artificial radiocarbon into the atmosphere makes it possible to investigate the distribution of the ^{14}C concentration in the growth rings in trees in the nuclear reactor deployment localities. Assumedly, ^{14}C enters the atmosphere with the nuclear reactor emission (largely in the form of $^{14}CO_2$), fits into the natural carbon cycle, and is absorbed by vegetation in the process of photosynthesis. It is expected that there will be more ^{14}C observed in the tree growth rings, the larger quantity of it entered the atmosphere in the given ring formation year, that is, the atmospheric concentration of ^{14}C in the woody plant growing period through the year will correlate with the concentration of ^{14}C in the growth ring for the given year.

The territory chosen to test the above assumptions was the Middle Urals which contains nuclear reactors of different types in operation at the Beloyarsk Nuclear Power Plant (BelNPP) and a research nuclear reactor operated by the Institute of Nuclear Materials (INM).

The parameters of the above reactor plants are presented in Table 1.

The amount of ^{14}C forming in nuclear reactors depends on the fuel enrichment, the concentrations of nitrogen impurities in fuel and structural materials, and the fuel assembly, coolant and moderator temperature.

Table 1. Parameters of nuclear reactors (The Power Reactor Information System)

Operation years	AMB-100 1964 – 1983	AMB-200 1969 – 1990	BN-600 from 1981	BN-800 from 2016	IVV-2M from 1966
Coolant	Light water	Light water	Liquid Na	Liquid Na	Light water
Moderator	Graphite	Graphite	–	–	Light water
Electric power, MW	102	160	560	820	15 (thermal)

The AMB-100 and AMB-200 nuclear plants are predecessors of the RBMK water-cooled graphite-moderated reactors, so reactions a) and c) are the key mechanisms for the ^{14}C formation. Since the BN-600 and BN-800 reactors are fast-neutron reactors, they do not have moderator. Therefore, the major sources of the ^{14}C formation will be oxygen in fuel and nitrogen impurities in fuel and fuel cladding, and the key ^{14}C formation mechanisms will be reactions a) and b). The IVV-2M nuclear research reactor is the prototype of the VVER water-cooled water-moderated reactor. It is used for production of radioisotopes (^{192}Ir, ^{14}C, ^{177}Lu, ^{131}Cs) and for all kinds of materials research (Russkikh 2017). The IVV-2M atmospheric emission contains ^{14}C formed largely by reactions a) and b), and as a result of handling the ^{14}C radioisotope part of which can enter the ventilation system.

The studies presented in the paper add to the findings on the subject matter of interest from studies by foreign authors. An analysis of the ^{14}C concentration in the components of environment in the vicinity of the Ignalina NPP, Lithuania, with two RBMK-1500 reactors is provided in (Magnusson et al. 2007, Mazeika et al. 2008, Ežerinskis et al. 2018). A similar study is presented in (Janovics et al. 2013) for the Paks NPP in Hungary which has four operating VVER-440 reactors. Specific to the latter is that it looks into the regularities of the ^{14}C distribution in the growth rings of a pine trees growing in the vicinity of a site with a number of different nuclear reactors.

Instrumentation and techniques

For the study, the location of the critical area was identified with the maximum radiation effects from the nuclear reactor release. The maximum bulk activity of ^{14}C in the air is assumed to be reached in the critical area. The calculation for the critical area was undertaken, as shown in (RB-106-15 2015), using data from the weather station of Verkhneye Dubrovo (25 km off the city of Zarechny). Common pine (Pinussylvestris L.), the most representative type of woody vegetation aged between 40 and 70 years, was chosen in the critical area as the target. Fig. 1 shows schematically the locations of the ^{14}C emission sources

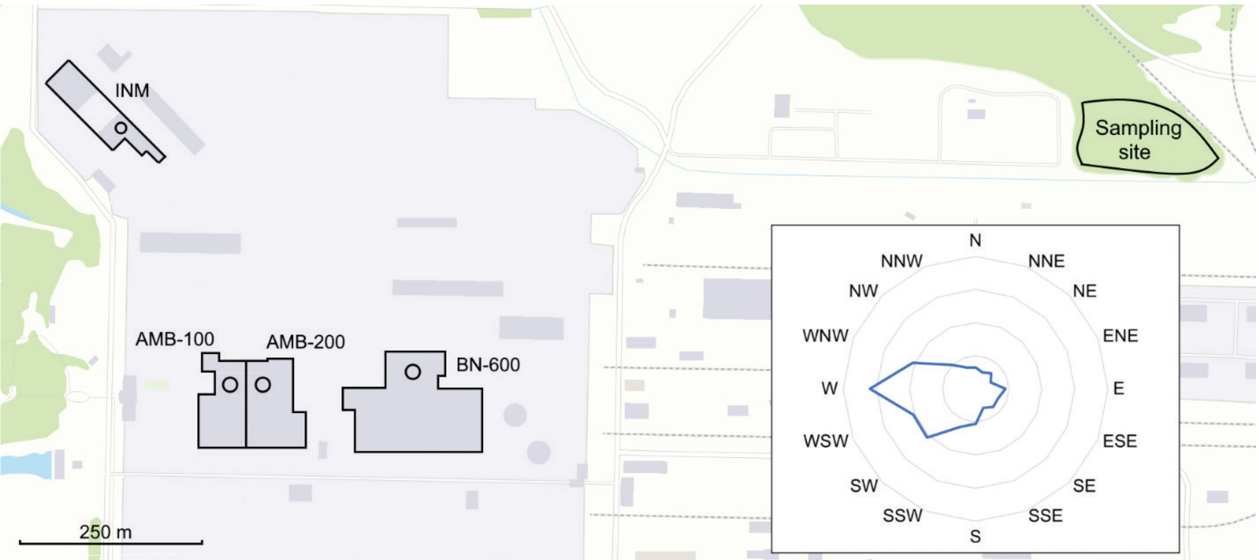

Figure 1. Locations of nuclear reactors and the emission area.

and the wood core sample taking point, as well as the wind rose with prevailing westerly and south-westerly winds.

The loads for identifying the ^{14}C content were formed by taking the wood core samples of the diameter 5 mm (Fig. 2), using an increment borer, at a height of about 130 cm from the soil surface. The cores collected were then separated into growth rings. The rings of one age from 10 specimens were combined as one sample for the respective year. There were 15 different samples chosen for the analysis.

Data on the ^{14}C activity in the growth rings of a 113-year-old pine tree in Akademgorodok, the city of Novosibirsk, was used as the background values. This is explained by two factors: first, Zarechny and Novosibirsk are situated approximately in the same latitude (55°02′ and 56°48′ northern latitude) and, second, Nobosibirsk is rather far from nuclear facilities both in operation and out of service, that is, has not been affected by anthropogenic radiocarbon sources, excluding nuclear weapon tests. In 2009, the specific activity of ^{14}C in land ecosystems in the northern hemisphere was 238 Bq/kg of C, which is close to the values prior to atmospheric nuclear tests (227 Bq/kg of C) (Carbon-14 2010).

Cellulose was chemically isolated as part of preparing the growth rings, which was further totally oxidized to form CO_2 and transformed into graphite-like carbon in a absorption-catalytic setup designed for producing AMS targets (Lysikov et al. 2018). The obtained targets were installed into an accelerator mass spectrometer and the concentration of ^{14}C was further measured.

The content of ^{14}C was analyzed as part of the study in 30 loads (two loads per year) using the accelerator mass spectrometer. Unlike other ^{14}C measurement techniques (Nazarov et al. 2021), a wood sample of about 20 mg is enough for the reliable AMS analysis which allows analyzing individual growth rings and even seasonal ring parts. The accelerator mass-spectrometer used was built in 2011 by the Budker Institute of Nuclear Physics of the Siberian Branch of the Russian Academy of Sciences in Novosibirsk. The operation of the device is described in detail in (Alinovsky et al. 2009).

Figure 2. Common pine core samples.

Results and discussion

The results of measuring the concentration of ^{14}C in the growth rings of ten common pine trees found in the critical locality area are presented in Fig. 3.

The ^{14}C measurement units (Percent Modern Carbon or pMC) were adopted in the second half of the 20th century. 100 pMC = 227 Bq/g of carbon is equivalent to the hypothetical specific activity of ^{14}C in the atmosphere in 1950 without human impacts (Stenstroom et al. 2011). The diagram shows a substantial growth in the concentration of ^{14}C in the mid-1960s, which is explained by radiocarbon entering intensively the atmosphere as a result of atmospheric nuclear weapon tests. The findings converge well with the literature data specific to the entire northern hemisphere (Levin et al. 1994). The maximum concentration of ^{14}C was 192.0 ± 8.5 pMC in 1964.

It can be seen in Fig. 3 that a growth was observed in the concentration of ^{14}C in the pine tree rings following the commissioning of the Beloyarsk NPP's units 1 and 2 with the AMB-100 and AMB-200 reactors. The concentration of radiocarbon in the atmospheric air was decre-

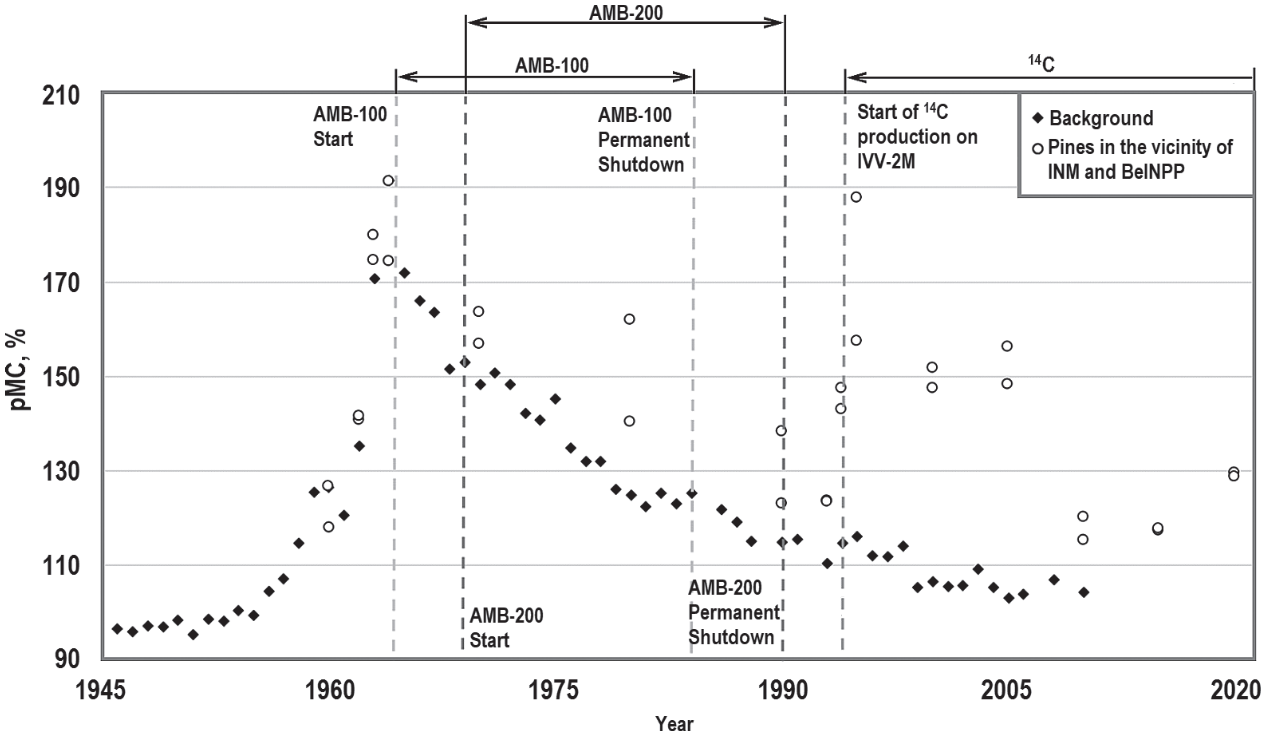

Figure 3. Results of measuring the concentration of 14C in the growth rings of pine trees in the IVV-2M, AMB-100 and AMB-200 reactor critical emission area. The dash lines show the boundaries of the reactor plant operating periods, and the dotted line shows the BN-600 startup.

asing up to the year 1993 as a result of decommissioning uranium-graphite reactors. The substantial differences in the concentration of ^{14}C during one year (shown by an example of 1985) can be explained by ^{14}C entering the atmosphere in a non-uniform manner during operation of the AMB-100 and AMB-200 nuclear reactors. Failures of fuel elements were likely to be behind the increased release of ^{14}C into the atmospheric air.

The data obtained proves indirectly the advantages of fast-neutron nuclear reactors as compared with uranium-graphite reactors in terms of environmental impacts from the release of ^{14}C they produce. The commissioning of the BN-600 fast-neutron reactor in 1980 had a minor effect on the concentration of ^{14}C in the atmospheric air. Unlike uranium-graphite nuclear plants, fast-neutron reactors have an order of magnitude smaller ^{14}C specific emission ratio (quantity of ^{14}C released into the atmosphere per unit of generated electricity): $1.4 \cdot 10^{-2}$ and $1.6 \cdot 10^{-1}$ GBq/GW·h respectively (Nazarov et al. 2018, IAEA-TECDOC-1638 2010).

After the AMB-100 and AMB-200 reactors stopped to operate, the key process leading, as a result, to the entry of ^{14}C into the atmosphere has been the isotope handling procedure begun at the INM in 1994. Reliable differences in the values of the ^{14}C concentration in the pine tree growth rings (specifically in 1995) are likely to be caused by the development of the methodology to handle ^{14}C as the raw material for radiopharmaceuticals. The subsequent decrease in the concentration of ^{14}C in the growth rings is explained by the improvement and optimization of the process.

Conclusions

The paper presents the results of measuring the concentration of ^{14}C in the growth rings of a pine tree from the critical area in the locality in the vicinity of the BelNPP and the INM. The measurements were perform using a unique scientific facility, the Accelerator Mass-Spectrometer of the Budker Institute of Nuclear Physics of the Siberian Branch of the Russian Academy of Sciences, based in Novosibirsk. The results of the ^{14}C activity observations in the growth rings of a 113-year-old pine tree growing outside the area affected by operating nuclear reactors and radiocarbon handling activities was used as the target for comparison.

In the tested pine tree samples, the concentration of ^{14}C in the growth rings is in a range of 116.0 ± 4.4 to 192.0 ± 8.5 pMC. The growth ring age determination allowed reproducing retrospectively the change in the levels of the ^{14}C release effects during the period from 1964 to present time. The ^{14}C impacts in the locality's critical area surveyed exceeded greatly the impacts in the background area compared against (year to year).

The additional anthropogenic entry of ^{14}C into wood was a result of the AMB-100 and AMB-200 nuclear plant operations. Following the decommissioning of the above reactors, the key source of the ^{14}C entry into the atmosphere was the INM's isotope handling activities and operation of the IVV-2M nuclear reactor. Operation of the BN-600 reactor does not contribute much to the concentration of ^{14}C in wood.

The accelerator mass spectrometry technique used to measure the ^{14}C concentration as part of the study is the most sensitive one among the currently available radiocarbon measurement techniques. Being used to address the nuclear industry objectives, this method will make it possible to obtain new precision data on the content of ^{14}C in components of the environment for estimating, retrospectively, the radiation impacts from nuclear facilities.

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