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^{14}C AND OTHER RADIONUCLIDES IN THE ENVIRONMENT IN THE BORDER REGION OF LITHUANIA BEFORE THE START OF THE BELARUSIAN NUCLEAR POWER PLANT OPERATION

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ABSTRACT. In this paper, we analyze the background activity of anthropogenic radionuclides (^{14}C, ^{3}H, ^{137}Cs, and ^{239,240}Pu), emphasizing ^{14}C content, in terrestrial and aquatic ecosystems in the Lithuanian border region before the commissioning of a new nuclear power plant in Belarus (BelNPP). In terrestrial samples, the ^{14}C concentration varied insignificantly—from 98.6 ± 0.7 to 102.2 ± 0.8 pMC, which is close to the ^{14}C level in atmospheric CO_2. In aquatic samples, the ^{14}C concentration varied within wide limits from 76.9 ± 0.7 to 99.6 ± 0.6 pMC, depending on the ecological group of macrophytes. Various ecological groups of macrophytes have experienced the influence of a freshwater reservoir effect. This lowest ^{14}C content in submerged macrophyte species, within the limits of uncertainty, was very close to the specific activity of ^{14}C in DIC (78.6 ± 0.6 pMC) in the water of the Neris River. The background ^{14}C values, together with the data on ^{3}H, ^{137}Cs and ^{239,240}Pu obtained in this study, can be used in the future to assess the contribution of the BelNPP conventional radioactive effluents to the levels of ^{14}C and other radionuclides in terrestrial and aquatic ecosystems of the transboundary region of Belarus and Lithuania.

KEYWORDS: ceasium-137 (^{137}Cs), nuclear power plants (NPPs), plutonium-239, 240 (^{239,240}Pu), radiocarbon (^{14}C), tritium (^{3}H).

INTRODUCTION

With the development of the global nuclear energy industry, the most serious accidents occurred at the Chernobyl nuclear power plant in April 1986 and at the Fukushima Daiichi nuclear power plant in March 2011 (Trapeznikov et al. 2007; Hirose 2012; Suchara 2017). These events have led to excessive public skepticism about plans to develop nuclear power in many countries. Today, such a situation exists in Lithuania, which, upon joining the European Union, shut down the only nuclear power plant in the country, the Ignalina NPP (INPP), and started to decommission it. The operation history of INPP, consisting of two reactor units, only lasted for 26 years. As a part of the obligations of the European Union Accession Treaty, Unit 1 of INPP was shut down on 31 December 2004, and Unit 2 on 31 December 2009. The history of the INPP operation and data on radiation in the environment indicate that the INPP was operated safely and benefited the society (https://www.iae.lt).

Active discussions in Lithuania on nuclear energy issues, both within the general public and on a political level, resumed again when the neighbouring country Belarus began to actively develop the projects for the construction of a nuclear power plant in the immediate proximity of the state border with Lithuania. Today, the Belarusian Nuclear Power Plant (BelNPP) has already been constructed, and the first unit was put into operation at the end of 2020. The industrial site of BelNPP is located close to the Lithuanian border (20 km) and the capital of the country, Vilnius (50 km). A part of the BelNPP 30-km zone is located on Lithuanian territory (Figure 1).

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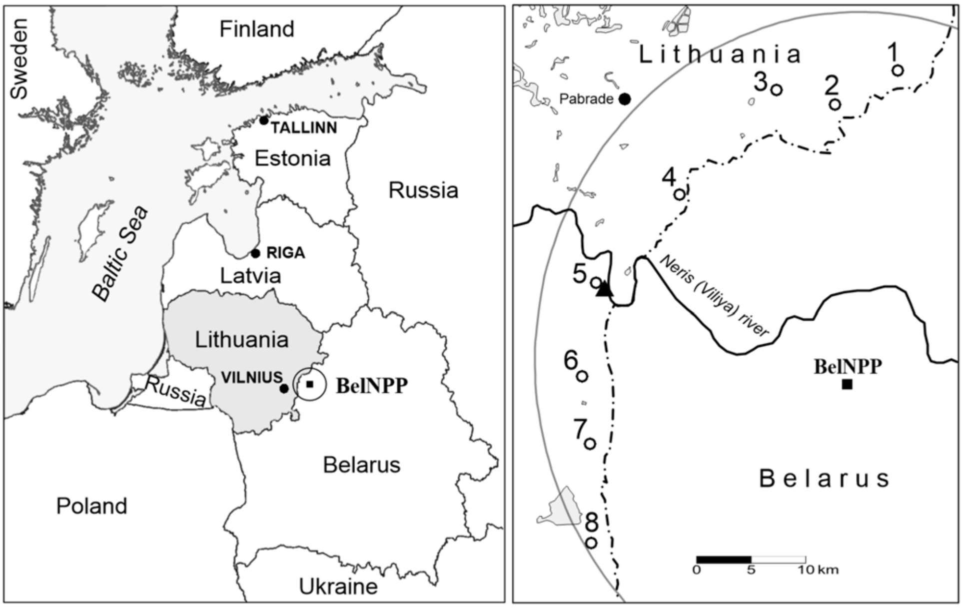

Figure 1 Sampling locations (a triangle in the scheme represents a position of surface water sampling point at Neris River, the circles represent the position of 8 monitoring sites for terrestrial ecosystems).

The increase in the number of operating nuclear power reactors in the east Baltic region and sensitive attitude to this issue from the general public facilitates the need for new research and the implementation of monitoring programs to keep the discussions on anthropogenic radionuclides originating from the BelNPP scientifically sound in future.

As it was previously shown in many studies (Mikhailov et al. 1999; Mažeika 2002; Nedveckaitė et al. 2007; Jasiulionis and Rozkov 2007; Gudelis et al. 2010; Mazeika et al. 2016; Jefanova et al. 2018), Lithuanian terrestrial and aquatic ecosystems near the INPP contained traces of carbon-14 (^{14}C), tritium (^{3}H), cobalt-60 (^{60}Co), and caesium-137 (^{137}Cs) originating from the INPP. We assume that the same radionuclides, especially mobile ones, such as ^{14}C and ^{3}H, may be present in the BelNPP environment in the future during its normal operation. Therefore, we initiated a baseline study of these radionuclides distribution in the Lithuanian part of the 30-km zone of the new NPP before the start of its operation. Some results on this topic have already been published. The aim of the study (Marčiulionienė et al. 2017) was to determine activities of ^{137}Cs and plutonium (Pu) isotopes and identify the sources of their origin in flooded soil and bottom sediments attributed to the water system of Neris and Nemunas Rivers and the Curonian Lagoon. The distribution of anthropogenic radionuclides (^{14}C, ^{3}H, ^{137}Cs, and ^{239,240}Pu) in forest soils and plants at the Lithuanian border region before the start of the BelNPP was analyzed by Jefanova et al. (2020).

The aim of this paper, therefore, is to present the distribution of ^{14}C in terrestrial and aquatic ecosystems at the Lithuanian border region before the start of the BelNPP operation. Because of the mobility, well-known atmospheric abundance, biological importance of carbon (^{12}C, ^{13}C, and ^{14}C), and the biological incorporation of radioactive ^{14}C through photosynthesis, it is of great importance to run ^{14}C measurements in the environment surrounding nuclear facilities. ^{3}H is the second mobile radionuclide that in addition to global origin can be traced in the environment surrounding nuclear facilities, especially in water bodies. The rather wide spreading of ^{3}H from NPPs is due to its transport with atmospheric precipitation and river runoff. Therefore, in this work, much attention is paid to the temporal distribution of ^{3}H in atmospheric precipitation and in the water of the Neris River, which will receive operational liquid discharges from the BelNPP. Furthermore, the data on other radionuclides (^{3}H, ^{137}Cs, and ^{239,240}Pu) are given here as evidence of the baseline pre-operational radioecological state of the area, but they are presented only briefly for comparison.

SAMPLING SITES, MATERIAL, AND METHODS

For a proper understanding of migration of the radionuclides upon release in the environment, the ecosystem components of interest and representative sampling sites must be carefully selected. Studies of the environmental consequences of the Chernobyl accident have evidenced that, for example, the uptake and retention of ^{137}Cs have generally been much higher in semi-natural ecosystems than in agricultural ecosystems (Balonov 2013). To represent terrestrial ecosystems in this study, the monitoring sites in forest ecosystems were established in the Lithuanian part of the 30-km zone of the BelNPP (Jefanova et al. 2020). The eight sampling sites were selected near the Belarus-Lithuania border in a ~60-km-long semi-regular arc (Figure 1). Forest ecosystems on *Arenosols* with a low groundwater table are spread in the Lithuanian part of 30-km zone territory of the BelNPP. The undisturbed organic soil is composed of the sequence of organic topsoil horizons, OL (organic litter), OF (organic fragmented horizon) and OH (organic humus horizon), distinguishable according to the decomposition degree of organic matter (IUSS 2015). Mineral topsoil horizon A composed of sand underlies by the organic horizons and contains $< 20\%$ of organic material (IUSS 2015). Blueberry pine forest more than 50 years old with rowan and other shrubs dominates here. The sampling sites are represented by natural pine forest ecosystems with low human impact.

A quadrat frame of 400 cm^{2} in area and 20 cm in height was used to collect organic and mineral soil samples for radionuclide analysis. The frame was driven vertically into the soil with hummer until reaching 20 cm in depth from the moss surface. The sampling point distance from the stems of trees was > 3 m. Series of living moss and soil horizons were collected.

Terrestrial plant samples included mosses and vascular plants: blueberry shrubs (*Vaccinium myrtillus* L.), rowan leaves (*Sorbus aucuparia* L.), mugwort stems (*Artemisia* sp.) and birch leaves (*Betula* sp.). *Pleurozium schreberi* prevailed in sampling sites 1 and 3–7, and *Hylocomium splendens* prevailed in sampling sites 2 and 8. In most cases, samples were mixes of different moss taxes.

To study the aquatic ecosystem under the influence of the BelNPP, a monitoring station (station 5, see Figure 1) was selected on the Neris River in immediate proximity of the state border with Belarus. Aquatic biota samples included: arrowhead (*Sagittaria* sp.), flowering rush (*Butomus umbellatus*), water-plantain (*Alisma* sp.), sedge (*Carex rostrata*), European bur-reed (*Sparganium* sp.), pondweed (*Potamogeton* sp.), common reed (*Phragmites australis*), frogbit (Hydrocharis morsus-ranae), Canadian waterweed (*Elodea canadensis*), and freshwater bivalve mussels (*Anadonta* sp. and *Unio* sp.).

Macrophytes were taken according to standard radioecological methods using a special hook (Marčiulionienė et al. 2017). Along with biota samples, bottom sediment and water samples were also taken for further analysis. The 5-cm-thick top layers of bottom sediment were collected using an Ekman bottom grab sampler (20 × 20 cm). Large volume water samples were collected using a submersible pump. To determine the activity of ^{14}C in water, dissolved inorganic carbon (DIC) was precipitated using appropriate quantities of NaOH and $CaCl_2$ as described in (Arslanov 1987). To determine the total activity of ^{137}Cs in water, including soluble and associated with particles, pretreated by ion exchange material filters were used (Lehto and Hou 2011).

A sampling of terrestrial and aquatic biota for all types of analysis, as well as water for the determination of ^{14}C and ^{137}Cs was carried out at the end of the growing season, namely in August or September of 2017–2020. Soil sampling for all types of analysis with a very limited number of samples for Pu isotopes was carried out once in 2017, with the exception of the soil profile for the ^{14}C, which was implemented in 2021.

At present, the level of ^{3}H in atmospheric precipitation is mainly due to its cosmogenic origin, with a pronounced minimum in winter and maximum in summer. Therefore, the dynamics of ^{3}H as one of the most mobile radionuclides in the aquatic environment, which can be influenced by the BelNPP, should be accurately compared with the variable background levels of ^{3}H in atmospheric precipitation. To take water samples from the Neris River to determine ^{3}H once a month, the same monitoring station 5 was used, from which samples were also taken for other analyses. Furthermore, for monthly precipitation collection for ^{3}H determination, two stations of east Lithuania were used: Zarasai (55°43'46"N, 26°10'42"E) and Vilnius (54°46'40"N, 25°17'36"E).

After standard pretreatment of corresponding material, the specific activity of ^{14}C in samples of plant, soil, and DIC in water was measured using the liquid scintillation counting (LSC) method (Gupta and Polach 1985; Arslanov 1987). A conventional procedure for benzene synthesis was applied (Kovaliukh and Skripkin 1994). ^{14}C activity counting in benzene was performed with a TriCarb 3170 TR/SL.

The specific activity of ^{3}H in monthly samples of atmospheric precipitation, river water periodic samples, and tissue free water tritium (TFWT) form of plant samples was measured using the low-background LSC method according to the procedure (ISO 9698 2019). The water fraction for ^{3}H determination was extracted from plant samples using the vacuum distillation and lyophilization methods. The precipitation and river water samples underwent primary distillation, electrolytic enrichment, neutralization and final distillation. Eight milliliters of tissue-free water or water after electrolytic enrichment were mixed with the scintillation cocktail, and ^{3}H activity was measured with a Quantulus 1220.

Air-dried terrestrial and aquatic plants, bivalve mussels, and samples from organic soil sub-horizons OL, OF and OH were combusted in a muffle furnace at 450°C for 5 hr. Deeper soil layers and crushed mussel valves were measured in the dry condition. Two geometries, 60 and 3 mL, were applied. Gamma-ray spectrometry using an ORTEC gamma-ray spectrometer with an HPGe GWL-120-15-LB-AWT detector (resolution 2.25 keV at 1.33 MeV) was applied to measure the specific activity of gamma-emitting radionuclides in biotic and soil samples (Gudelis et al. 2000; Jefanova et al. 2020).

In order to determine the specific activity of plutonium isotopes, the ash samples of plants, bottom sediment and soil were dissolved in strong acids (HNO_3, HCl, HF, and $HClO_4$). TOPO/cyclohexane extraction and radiochemical purification using TEVA resins (100–150

Table 1 ^{14}C specific activity (pMC, ± 1 sigma/Bq/kg of biota*, dry weight, ± 1 sigma) in terrestrial plants (mugwort, *Artemisia vulgaris*) from Lithuanian part of the 30 km zone of the BelNPP, Belarus.

	Year		
Site no.	2018	2019	2020
1	99.1 ± 0.8/124.5 ± 1.0	99.9 ± 0.6/125.5 ± 0.8	99.8 ± 0.6/125.3 ± 0.8
2	100.0 ± 0.8/125.6 ± 1.0	100.1 ± 0.7/125.7 ± 0.9	100.2 ± 0.6/125.8 ± 0.8
3	100.8 ± 0.8/126.6 ± 1.0	98.6 ± 0.7/123.8 ± 0.9	99.7 ± 0.6/125.2 ± 0.8
4	99.5 ± 0.8/125.0 ± 1.0	99.8 ± 0.6/125.3 ± 0.8	100.0 ± 0.6/125.6 ± 0.8
5	99.1 ± 0.9/124.5 ± 1.1	101.5 ± 0.7/127.5 ± 0.9	100.8 ± 0.6/126.6 ± 0.8
6	100.8 ± 0.8/126.6 ± 1.0	100.5 ± 0.7/126.2 ± 0.9	101.3 ± 0.6/127.2 ± 0.8
7	99.8 ± 0.8/125.3 ± 1.0	101.6 ± 0.7/127.6 ± 0.9	100.4 ± 0.7/126.1 ± 0.9
8	100.2 ± 0.8/125.8 ± 1.0	102.2 ± 0.8/128.4 ± 1.0	100.1 ± 0.8/125.7 ± 1.0

*Calculated basing on approximate C_{org} fraction in biota material.

μm) were used to separate Pu isotopes. Pu isotopes were electroplated onto stainless steel disks and measured using an alpha-spectrometry system with passivated implanted planar silicon (PIPS) detectors with an active area of 450 mm^2 (AMETEK, Oak Ridge, Tennessee, USA). For more information, see Lujanienė (2013).

RESULTS AND DISCUSSION

The data on ^{14}C specific activity in terrestrial plants (mugwort) in the Lithuanian part of the BelNPP 30-km zone for the period of observations 2018–2020 are presented in detail in Table 1.

We compared the data on ^{14}C for annual terrestrial plant species from the edge of the 30-km zone of the BelNPP with the data on the level of ^{14}C in atmospheric CO_2, which is currently approaching the level of ^{14}C from cosmogenic origin ~100 pMC. The background $^{14}CO_2$ levels due to fossil fuel burning CO_2 emissions depending on reduction scenarios are predicted to reach pre-1950 levels (i.e., $^{14}CO_2 < 100$ pMC) by 2021 in the Northern Hemisphere with a 20% probability (Sierra 2018; Zhang et al. 2021).

^{14}C specific activity in mugwort from all sites studied in the BelNPP 30-km zone varied insignificantly (Table 2). In all samples for 2018–2020, the specific activity of ^{14}C varied from 98.6 ± 0.7 to 102.2 ± 0.8 pMC, and the mean value within standard deviation (SD) was 100.2 ± 0.9 pMC, which is close to the current level of ^{14}C in the atmosphere (Hua et al. 2021). The average value of ^{14}C specific activity in mugwort (± SD) for different years was as follows: 99.9 ± 0.7 pMC in 2018, 100.5 ± 1.2 pMC in 2019, and 100.3 ± 0.6 in 2020. All these data sets do not differ statistically significantly, and the average value of ^{14}C specific activity in mugwort can be taken for the terrestrial ecosystem as a ^{14}C background value. A slight depletion of ^{14}C in some plant samples (up to 1 pMC) relative to atmospheric CO_2 can be associated with the peculiarities of plant metabolism or the input of a small fraction of “old” carbon. The ^{14}C background value can be used in the future when assessing the contribution of routine airborne emissions from BelNPP to the ^{14}C level in the terrestrial ecosystem.

For example, we refer to the ^{14}C data attributed to the terrestrial environment of the INPP and collected throughout the entire period of its operation. These data recorded the

Table 2 ^{14}C specific activity in living moss and soil horizons taken from pine forest ecosystem in Lithuanian part of the 30 km zone of the BelNPP, Belarus (sampling point 8, see Figure 1).

Soil profile components	Thickness, cm (Corg, %)**	^{14}C (± 1 sigma), pMC	Calibrated age*, AD (main range of 95.4% confidence intervals)
Living moss	3 (58.2)	99.12 ± 0.67	n/e***
OL (organic litter)	2 (57.9)	100.74 ± 0.54	2014 – … (69.5%)
OF (organic fragmented horizon)	2 (46.5)	105.50 ± 0.56	2005 – 2011 (87.3%)
OH (organic humus horizon)	1 (16.8)	109.45 ± 0.75	1997 – 2002 (83.3%)
Horizon A1	1 (3.7)	109.40 ± 0.88	1996 – 2003 (83.3%)
Horizon A2	5 (3.0)	108.37 ± 1.39	1996 – 2008 (84.1%)

*Calibrated using OxCal 4.4 software (Bronk Ramsey, 2009) and post-bomb atmospheric NH1 curve (Hua et al. 2021).
**Corg derived from loss on ignition (LOI) data.
***Data does not fit post-bomb atmospheric NH1 curve.

^{14}C activity excess compared to the corresponding ^{14}C background level of certain periods. Pine tree ring measurements evidenced an increase in ^{14}C concentrations by 3–6 pMC during the first 14 years of INPP operation (1983–1997) when no maintenance works of the reactors were needed (Mazeika et al. 2008). During the operational period of 1998–2003, increased ^{14}C specific activity values up to 6–14 pMC coincided with the replacement events of the zirconium alloy tubes of the fuel channels in both units of the INPP (Ežerinskis et al. 2018).

The data on ^{14}C in the soil profile in the Lithuanian part of the BelNPP 30-km zone for the recent period of observations (2021) are presented in detail in Table 2.

The content of ^{14}C in living moss (99.1 ± 0.7 pMC) did not differ from its content in other terrestrial plants, as well as in organic litter (100.7 ± 0.5 pMC). The content of ^{14}C in organic soil horizons varied more considerably compared to that of living moss and organic litter depending on the mean time elapsed since carbon in the soil system was fixed from atmospheric CO_2. Soil organics age evaluation using OxCal 4.4 software (Bronk Ramsey, 2009) and post-bomb atmospheric NH1 curve (Hua et al., 2021) revealed the following values: living moss and OL horizon < 10 yr, OF and OH horizons: 13 ± 3 and 22 ± 3 yr, respectively. The age of organics admixture in the sand (horizons A1 and A2) was similar to that of organic soil horizons (Table 2).

The data on ^{3}H activity in terrestrial plants and atmospheric precipitation within the 30-km zone of the BelNPP for the period of observations (2018–2020) are presented in Table 3 and Figure 2.

The TFWT values in rowan leaves selected from the 30-km zone of the BelNPP in different years varied insignificantly (averaged value ±SD): 8.8 ± 1.0 TU for samples taken on 08-08-2018, 6.1 ± 1.6 TU – on 20-09-2019, 9.2 ± 1.1 TU – on 17-08-2020. TFWT values for plants were close to the ^{3}H level in precipitation (Figure 2). The ^{3}H specific activity in monthly precipitation collected from Vilnius station was as follows: 11.4 ± 0.8 TU for August of 2018, 7.1 ± 0.5 TU for September of 2019 and 9.0 ± 0.6 TU for August of 2020.

Table 3 ^{3}H specific activity in tissue free water form (TU, ± 1 sigma/Bq/kg* of fresh biota, ± 1 sigma) of terrestrial plants (rowan leaves, *Sorbus aucuparia*) taken from Lithuanian part of the 30 km zone of the BelNPP, Belarus.

	Date		
Site no.	09-08-2018	20-09-2019	17-08-2020
1	9.2 ± 2.2/0.59 ± 0.14	<2.8/<0.18	9.5 ± 2.6/0.61 ± 0.17
2	8.7 ± 2.2/0.58 ± 0.15	4.1 ± 3.2/0.27 ± 0.21	8.7 ± 2.6/0.58 ± 0.17
3	7.3 ± 2.2/0.48 ± 0.14	6.0 ± 2.8/0.39 ± 0.18	9.3 ± 2.7/0.61 ± 0.18
4	10.2 ± 2.3/0.62 ± 0.14	5.1 ± 3.2/0.31 ± 0.19	8.1 ± 2.6/0.49 ± 0.16
5	8.0 ± 2.2/0.51 ± 0.14	8.2 ± 3.2/0.53 ± 0.21	9.7 ± 2.6/0.62 ± 0.17
6	10.3 ± 2.3/0.72 ± 0.16	8.3 ± 3.2/0.58 ± 0.22	10.8 ± 2.7/0.75 ± 0.19
7	8.2 ± 2.2/0.55 ± 0.15	5.0 ± 2.8/0.34 ± 0.19	7.4 ± 2.6/0.50 ± 0.17
8	8.8 ± 2.2/0.53 ± 0.13	6.3 ± 2.8/0.38 ± 0.17	10.2 ± 2.7/0.62 ± 0.16
In monthly precipitation (TU/Bq/L)	11.4 ± 0.8/1.3 ± 0.1	7.1 ± 0.5/0.8 ± 0.1	9.0 ± 0.6/1.1 ± 0.1

*Calculated basing on tissue free water content of terrestrial plants.

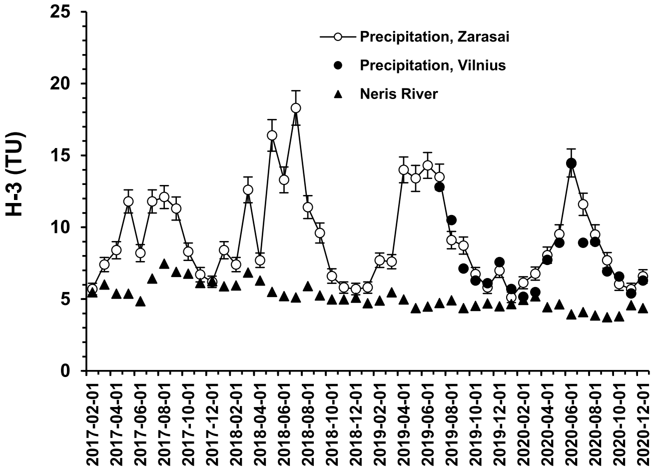

Figure 2 ^{3}H activity concentration in monthly atmospheric precipitation in eastern Lithuania. The observations in long-term monitoring station Zarasai in 1999 (in detail after Jefanova et al. 2018) and in Vilnius station in 2017 were started. There is a strong correlation between ^{3}H from two precipitation stations ($y = 0.9947x - 0.0616$, $R^2 = 0.81$). Together ^{3}H activity concentration in Neris River water sampled one time a month is shown.

Despite its low radiation significance, ^{3}H is a very important specific mobile radionuclide, due to its complex global inventory from thermonuclear weapons testing and from cosmogenic production, as well as because of its local excess often being traced as originating from NPP sites (Jefanova et al. 2018). ^{3}H in atmospheric precipitation has been measured in Lithuania for the past 20 years, which allows the ^{3}H time series of monthly precipitation since 1999 to act as a basis for interpretation of the ^{3}H distribution in terrestrial and aquatic ecosystems, both for background areas and NPP sites.

Table 4 ^{137}Cs specific activity (Bq/kg dry weight ± 1 sigma) of terrestrial plants (rowan leaves, *Sorbus aucuparia*) taken from Lithuanian part of the 30 km zone of the BelNPP, Belarus.

	Year				
Site no.	2017*	2017*	2018	2019	2020
1	7.2 ± 0.7/1010 ± 105**	7.5 ± 0.6	7.5 ± 0.5	3.7 ± 1.4	5.8 ± 0.4
2	17.1 ± 0.7/930 ± 35	n/s***	12.0 ± 0.7	5.3 ± 1.5	1.1 ± 0.5
3	46.7 ± 1.4/1650 ± 215	8.9 ± 0.4	6.9 ± 0.5	1.3 ± 0.4	3.1 ± 0.4
4	43.5 ± 1.2/1600 ± 140	4.8 ± 0.4	3.5 ± 0.4	3.5 ± 0.6	3.8 ± 0.4
5	11.4 ± 1.4/1230 ± 150	2.8 ± 0.6	1.1 ± 0.3	6.5 ± 0.9	4.1 ± 0.5
6	27.3 ± 1.2/1110 ± 135	7.5 ± 0.6	5.3 ± 0.6	7.9 ± 0.8	6.1 ± 0.3
7	50.2 ± 1.3/1470 ± 155	2.8 ± 0.3	10.1 ± 0.8	30.8 ± 0.9	9.6 ± 0.4
8	39.5 ± 1.1/1410 ± 180	18.9 ± 0.8	10.3 ± 0.6	11.4 ± 1.0	9.7 ± 0.4

*After Jefanova et al. (2020).
**^{137}Cs specific activity of soil Oh horizon (Bq/kg dry weight ± 1 sigma)/inventory of ^{137}Cs in soil (Bq/m^2 ± 1 sigma).
***n/s, not sampled.

The observed ^{3}H variations in precipitation of Zarasai station are similar to those at the GNIP station Vienna, Hohe Warte (Jefanova et al. 2018). ^{3}H monthly data clearly express seasonal variations with maximum values in the spring-summer months (May–August) and with minimum values in autumn-winter months (October–February). The averaged trend evidences the decline of ^{3}H from thermonuclear weapons testing to almost the ^{3}H level in precipitation corresponding to cosmogenic production. Very similar ^{3}H variations were observed since 2017 in Vilnius station, which is located closer to the BelNPP site compared to Zarasai station. However, both ^{3}H datasets are very similar with a correlation coefficient of 0.9 for the 2017–2020 period. The seasonal variations of ^{3}H activity in precipitation, with some smoothing and averaging effects dependent on the water turnover rate, form the ^{3}H background level in terrestrial plants.

Data on the specific activity of ^{137}Cs in terrestrial plants (rowan leaves) and soils in the Lithuanian part of the 30-km zone of the BelNPP for the last observation period (2017–2020) are presented in Table 4.

The average value of ^{137}Cs specific activity in rowan leaves (± SD) in different years was as follows: 7.6 ± 5.5 Bq/kg in 2017, 7.1 ± 3.7 Bq/kg in 2018, 8.8 ± 9.4 Bq/kg in 2019, and 5.4 ± 3.0 Bq/kg in 2020. The temporal dynamics of ^{137}Cs specific activity in rowan leaves at the monitoring points was mostly uniform over 4 years. Despite the fact that the average level of ^{137}Cs in the rowan leaves was low, it is characterized by a large areal variation in site 8 in 2017 and in site 7 in 2019. Individual cases of increased activity of ^{137}Cs up to 30 Bq/kg and even more may be associated with the resuspension of ^{137}Cs from soils, especially of agricultural land, and its airborne transport due to forest fires in Belarus and Ukraine that took place in the summer of 2019.

Analyzing briefly the data on ^{137}Cs specific activity in rowan leaves, one should pay attention to the ^{137}Cs activity changes at different sampling sites with time, as well as to spatial changes that depend on the areal distribution of ^{137}Cs inventory. The data on the specific activity of ^{137}Cs in typical soil profiles and various terrestrial plants have been studied in detail previously (Jefanova et al. 2020). One of the main findings of the study (Jefanova et al.

Table 5 ^{14}C specific activity in macrophytes and surface water samples taken from Neris River in Lithuanian part of the 30 km zone of the BelNPP, Belarus (sampling point 5, see Figure 1).

Macrophytes	Year	^{14}C (± 1 sigma), pMC	^{14}C (± 1 sigma) Bq/kg of biota, dry weight*, Bq/m^3 of water**
Arrowhead (*Sagittaria* sp.), emerged plants	2018	95.09 ± 0.92	74.9 ± 0.7
Flowering rush (*Butomus umbellatus*), emerged plants	2018	99.61 ± 0.65	76.3 ± 0.6
Frogbit (*Hydrocharis morsus-ranae*), floating plants	2018	96.29 ± 0.65	78.8 ± 0.5
Pondweed (*Potamogeton* sp.), submerged plants	2018	78.39 ± 0.57	58.9 ± 0.4
Canadian waterweed (*Elodea canadensis*), submerged plants	2018	76.88 ± 0.67	57.8 ± 0.5
Lesser water-plantains (*Baldellia* sp.), submerged plants	2018	78.50 ± 0.87	59.0 ± 0.7
^{14}C in dissolved inorganic carbon (DIC)	2018	78.0 ± 0.9	8.9 ± 0.2
	2019	78.8 ± 0.5	9.0 ± 0.2
	2020	79.1 ± 1.2	8.6 ± 0.2

*Calculated basing on approximate C_{org} fraction in biota material.

**Calculated basing on concentration of DIC in water.

2020) was the recovering of changes in ^{137}Cs inventory in the north-south direction as follows: 970 ± 110 Bq/m^2 in sampling sites 1 and 2, 1625 ± 260 Bq/m^2 in sampling sites 3 and 4, 1170 ± 200 Bq/m^2 in sampling sites 5 and 6 and 1440 ± 240 Bq/m^2 in sampling sites 7 and 8. Furthermore, natural fallout radionuclides (^{7}Be and ^{210}Pb) were found in rowan leaves, which indicate that rowan leaves effectively intercept atmospheric deposition during one growing season, and the contribution by roots is negligible. This fact makes it possible to use rowan leaves as a bioindicator of short-term airborne transport (year by year) of ^{137}Cs from a nuclear source.

We also found plutonium isotopes in soil samples with specific activity approaching ~1 Bq/kg (site 2, OF horizon 0.85 ± 0.04 Bq/kg; site 3, OH horizon 1.01 ± 0.10 Bq/kg; site 7, OH horizon 0.95 ± 0.05 Bq/kg) and an activity ratio of ^{238}Pu/^{239,240}Pu, a ratio typical of global fallout. Contrarily, lower activity concentration (by twice) containing traces of the Chernobyl-derived plutonium was found in a moss sample (site 7, 0.40 ± 0.04 Bq/kg).

Data on ^{14}C in macrophytes and surface water samples in the Lithuanian part of the 30-km zone of the BelNPP for the observation period 2018–2020 are detailed in Table 5.

The data on ^{14}C in macrophytes showed significant variability depending on the ecological group of macrophytes: from 76.88 ± 0.67 pMC in Canadian waterweed to 99.61 ± 0.65 pMC in a flowering rush. Different ecological groups of macrophytes are influenced to a varying degree by the so-called freshwater reservoir effect (FRE). This effect, known for many lakes and rivers (Philippsen 2013), is determined by the presence of dissolved ancient carbonates in the watershed area.

Floating macrophytes (*Hydrocharis morsus-ranae*) and habituating nearshore zone emerged plants (*Sagittaria* sp., *Butomus umbellatus*) had a very similar, compared to atmospheric CO_2 only slightly depleted ^{14}C content (~3 pMC): the average value ± SD was 97.0 ± 2.3 pMC. The submerged species (*Potamogeton* sp., *Elodea canadensis* and *Baldellia* sp.) were highly depleted with respect to ^{14}C level in the atmosphere (~22 pMC): the average value of ^{14}C specific activity ± SD was 77.9 ± 0.9 pMC. This value, within the limits of uncertainty, is very close to the specific activity of ^{14}C in DIC (78.6 ± 0.6 pMC), which indicates that submerged macrophytes utilize the carbon of DIC.

To show the radioecological significance of the background ^{14}C level, we calculated the ^{14}C activity concentrations in the samples in Bq units using the Corg content in biota (33–35% for macrophytes of the Neris River) and the DIC content in water (240–280 mg/L in the water of Neris River). Thus, the following levels of ^{14}C were obtained: 74.9–78.8 Bq/kg in emerged and floating plants, 57.8–59.0 Bq/kg in submerged plants, 8.6–9.0 Bq/m^3 in water.

The specific activity of ^{3}H in the water of Neris River was relatively constant with an average value ± SD of 5.9 ± 0.9 TU (Figure 2), which corresponds to low specific activity of ^{3}H in the fall-winter precipitation, which forms the base flow to the riverbed. This level of ^{3}H serves as a baseline for interpreting the distribution of ^{3}H excess in the aquatic ecosystem, which in the future may be affected by the BelNPP.

As in the case of ^{14}C, the distribution of ^{137}Cs in various environmental samples from the aquatic ecosystem of the Neris River was studied with similar detail (Table 6, Figure 3).

^{137}Cs and plutonium isotopes are of great concern among anthropogenic radionuclides prevalent in the environment after nuclear weapons testing and nuclear accidents at Chernobyl and Fukushima NPPs. From the river catchment area, a small fraction of the inventory of low-mobile radionuclides (^{137}Cs and Pu isotopes) is constantly transferred to the river ecosystem, where it is distributed between water, suspended matter, riverbed sediments and biota (Trapeznikov et al. 2007).

Since the watershed of the Neris River was slightly contaminated with artificial radionuclides as a result of global fallout and the fallout after the Chernobyl accident (Atlas 1995), the content of ^{137}Cs in the components of the aquatic ecosystem is low and fluctuates within the following ranges: from < MDA (minimum detectable activity) to 14.2 Bq/kg for macrophytes and from 1.2 to 1.9 Bq/kg for soft tissues of freshwater bivalve mussels. The above-ground part of macrophytes contains fewer ^{137}Cs than the roots: the average values of the specific activity of ^{137}Cs (± SD) were 5.4 ± 1.8 Bq/kg and 9.3 ± 4.5 Bq/kg, respectively. ^{137}Cs specific activity in valves of mussels was <MDA in all cases. In addition to biotic components, ^{137}Cs was also detected in riverbed sediments and water of the Neris River, where its specific activity ± SD was 5.8 ± 1.1 Bq/kg and 0.78 ± 0.43 Bq/m^3, respectively.

Furthermore, in riverbed sediments, the specific activity of $^{239,240}Pu$ was also determined, which was very low and varied in the range of 0.035–0.090 Bq/kg, and the average activity with a standard deviation was 0.057 ± 0.023 Bq/kg. $^{239,240}Pu$ in riverbed sediments from the Neris River at an observation site close to the state border with Belarus was mainly due to global fallouts after nuclear tests with minor traces of plutonium of Chernobyl origin in some places.

Table 6 ^{137}Cs specific activity in macrophytes, freshwater bivalve mussels, riverbed sediments and surface water samples taken from Neris River in Lithuanian part of the 30 km zone of the BelNPP, Belarus (sampling point 5, see Figure 1).

Material	Sample feature	Year	^{137}Cs (± 1 sigma) Bq/kg of biota, dry weight
Arrowhead (*Sagittaria* sp.)	Above ground part	2019	6.4 ± 2.3
	Roots		14.2 ± 2.5
Flowering rush (*Butomus umbellatus*)	Above ground part	2019	3.6 ± 1.0
	Roots		7.8 ± 1.0
Water-plantain (*Alisma* sp.)	Above ground part	2019	7.5 ± 2.1
	Roots		11.4 ± 1.4
Sedge (*Carex rostrata*)	Above ground part	2019	4.2 ± 1.3
	Roots		10.8 ± 1.4
European bur-reed (*Sparganium* sp.)	Not separated	2019	9.4 ± 1.9
Pondweed (*Potamogeton* sp.)	Above ground part	2019	<MDA (3.8)
Common reed (*Phragmites australis*)	Above ground part	2019	<MDA (3.2)
	Roots		2.3 ± 0.3
Bivalve mussels (*Anadonta* sp.)	Soft tissues	2019	1.3 ± 0.4
	Valves		<MDA (0.4)
Bivalve mussels (*Anadonta anatina*)	Soft tissues	2019	3.3±1.1
	Valves		<MDA (0.4)
Bivalve mussels (*Unio* sp.)	Soft tissues	2019	1.8 ± 0.5
	Valves		<MDA (0.4)
Bivalve mussels (*Unio pictorum*)	Soft tissues	2019	1.2 ± 0.3
	Valves		<MAD (0.5)
Bivalve mussels (*Unio tumidus*)	Soft tissues	2019	1.9 ± 0.5
	Valves		<MDA (0.5)
Riverbed sediment (muddy sand)	0–5 cm	2019	5.8 ± 0.6
River water (Bq/m^3)	Soluble and associated with total suspended matter	2018	0.34 ± 0.08
		2019	0.81 ± 0.06
		2020	1.20 ± 0.05

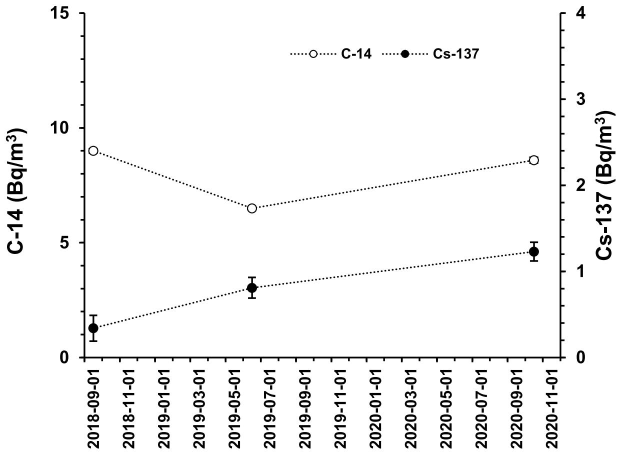

Figure 3 ^{14}C activity concentration in DIC and ^{137}Cs activity concentration (soluble and associated with total suspended matter) in water in Neris River.

CONCLUSIONS

We have collected data to characterize the main radioecological parameters of terrestrial and aquatic ecosystems located in the Lithuanian part of the 30-km zone of the BelNPP, as a basis for continuing the radioecological assessment when the BelNPP is operational. In the samples of terrestrial plants, ^{14}C concentration varied insignificantly from 98.6 ± 0.7 to 102.2 ± 0.8 pMC, which was close to the level of ^{14}C in atmospheric CO_2. The specific activity of ^{3}H in the TFWT form ranged from 6.1 ± 1.6 to 9.2 ± 1.1 TU, which corresponded to the ^{3}H level in precipitation for this region in the fall season. The relatively low ^{137}Cs inventory in this region determined the level of ^{137}Cs activity in terrestrial plants ranging from 1.0 ± 0.5 to 40.5 ± 1.8 Bq/kg dry weight. An increase in ^{137}Cs activity up to 30–40 Bq/kg and even more may be associated with the resuspension of ^{137}Cs from soils and forest fires in Belarus and Ukraine. We also found plutonium isotopes in soil samples with specific activity approaching ~1 Bq/kg.

In the samples of aquatic plants, the ^{14}C concentration varied within wide limits from 76.9 ± 0.7 to 99.6 ± 0.6 pMC, depending on the ecological group of macrophytes, which, to varying degrees, have experienced the influence of a freshwater reservoir effect. The specific activity of ^{3}H in the water of Neris River was relatively constant with an average value ± SD of 5.9 ± 0.9 TU, which corresponded to low specific activity of ^{3}H in the fall-winter precipitation. The content of ^{137}Cs in the components of the aquatic ecosystem was low and fluctuated from $<$ MDA to 14.2 Bq/kg for macrophytes and from 1.2 to 1.9 Bq/kg for soft tissues of freshwater bivalve mussels. ^{137}Cs was also detected in riverbed sediments and water of the Neris River, where its specific activity was 5.8 ± 1.1 Bq/kg and 0.78 ± 0.43 Bq/m^3, respectively. In riverbed sediments, the specific activity of ^{239,240}Pu was very low and varied in the range of 0.035–0.090 Bq/kg.

The data on artificial radionuclides (^{14}C, ^{3}H, ^{137}Cs, and ^{239,240}Pu) obtained in this study will be used in the future to assess the contribution of conventional radioactive effluents from BelNPP to the radionuclides level in terrestrial and aquatic ecosystems of the transboundary region of Belarus and Lithuania.

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