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ASSESSMENT OF THE CONTAMINATION BY ^{14}C AIRBORNE RELEASES IN THE VICINITY OF THE IGNALINA NUCLEAR POWER PLANT

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ABSTRACT. A radiocarbon (^{14}C) activity analysis in the tree rings around Ignalina nuclear power plant (INPP) has been carried out with the aim to test the hypothesis to use ^{14}C tree-ring analysis data as a tool for the reconstruction of gaseous releases from NPP to the environment. The INPP has been in decommissioning state since the end of 2009. Tree-ring samples for ^{14}C analysis were collected 7 yr after final power unit shutdown from the INPP vicinity. The samples from 5 sampling locations were collected, prepared and measured using the Single Stage Accelerator Mass Spectrometer (SSAMS). Data analysis represents observable Ignalina NPP influence by ^{14}C increase up to 15 pMC (percent modern carbon) in tree rings. Good correlations of the ^{14}C concentrations and wind direction were obtained. The main purpose of this article was to match ^{14}C measurement data along with the atmospheric dispersion modeling of emissions in order to retrospectively characterize the emission source.

KEYWORDS: airborne release, AMS, ^{14}C, correlation, Gaussian model, Ignalina NPP, tree rings, wind rose.

INTRODUCTION

The analysis of radiocarbon (^{14}C) activity in the environment reveals its changes, which can be correlated with the influence of the natural carbon cycle processes and anthropogenic activity. ^{14}C is produced in upper layers of the atmosphere by cosmic rays activating mainly the nitrogen atoms. Production of atmospheric ^{14}C is counteracted with slow removal by the radioactive decay (half-life is 5730 yr) and rapid processes of global carbon cycle due to the CO_2 exchange. CO_2 is removed by the oceans due to chemical transformation into the carbonates and by the capture and incorporation into organic materials of the $^{14}CO_2$ by the autotrophic organisms, mainly plants. These processes are responsible for the fast removal of $^{14}CO_2$ excess (Stuiver and Polach 1977; Stuiver 1983) from the atmosphere after the nuclear weapon tests of 1950s and 1960s. One remarkable mechanism of ^{14}C activity (per gram of C) reduction in the atmosphere is the Suess effect, which is the dilution of the ^{14}C due to admixture of ^{14}C-free CO_2 from fossil fuel burning installations, mainly transport and industry (Suess 1955). This phenomenon can be used to detect the CO_2 emissions of the fossil fuel origin coming from thermal power plants or, e.g., leakages of carbon capture and storage (CCS) installations (Turnbull et al. 2017).

Nuclear power plants (NPP) are also sources of ^{14}C release to the environment, along with other radionuclides, mostly of noble gasses, iodine, tritium. The ^{14}C in nuclear reactors is generated mainly by neutron reactions with ^{17}O, ^{14}N and ^{13}C isotopes present in the reactor core, moderator and coolant. The main source of ^{14}C in light water reactors is the coolant and graphite moderator in High temperature gas-cooled (HTGR), Advanced gas-cooled (AGR), Magnox and High power channel-type reactors (RBMK) (Kunz 1985; Yim and Caron 2006). From pressured water reactors (PWR), the ^{14}C is released mainly in the chemical form of hydrocarbons, such as methane or ethane, as well as carbon dioxide (13–30%) (Veres et al. 1995; Van der Stricht and Janssens 2001; Povinec et al. 2008). In boiling water reactors (BWR) and reactors with graphite moderator (HTGR, AGR, Magnox, RBMK) dominant ^{14}C releases are in the form of carbon dioxide (Gaiko et al.

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1985; Stenström et al. 2000; Yim and Caron 2006; Mažeika et al. 2008). Once generated, the gaseous chemical compounds containing ^{14}C are released to the environment either through special venting equipment during the operation or during reactor shutdowns. In particular, RBMK type reactors have ejectors for the separation and release of gaseous products from the turbine condensers to the ventilation system and finally to the atmosphere (Remeikis et al. 2012). Although the ventilation system of the RBMK reactors is equipped with special filters (aerosol, activated carbon, zeolite absorber and mechanical filters), some fraction of the ^{14}C is released to the environment. e.g., the total ^{14}C release to the environment was $2.2{\cdot}10^{10}$ Bq for the year 2008, 80% of this was released from the Unit 2 reactor building 101/2 as an airborne discharge when Unit 2 was still in operation (VATESI 2009).

The release of ^{14}C can be confirmed by tree-ring analysis of the samples collected from the vicinity of a NPP, as the slight increase of $^{14}CO_2$ concentration in the air is recorded as a pattern in the tree rings due to photosynthesis process and incorporation of $^{14}CO_2$ into cellulose matrix of tree wood. The airborne releases are responsible for the slight increase of the ^{14}C concentration, which can be detected with modern measurement equipment, mainly by accelerator mass spectrometry (AMS). E.g., in Žlkovce (monitoring station situated 5 km away from Bohunice NPP) $\Delta^{14}C$ was measured up to 300‰ (130 pMC), however near Dukovany NPP (Chech Republic) this was negligible resulting in 5‰ (0.5 pMC) increase in $\Delta^{14}C$ (Svetlik et al. 2012; Povinec et al. 2015). A slight increase was also observed at Barseback nuclear power plant (Sweden) and the vicinity of Paks nuclear power plant (Hungary) (Stenström et al. 1998; Janovics et al. 2017).

In our previous study (Ežerinskis et al. 2018), ^{14}C activity was analysed in the tree rings from one site close to the INPP located in northeastern part of Lithuania (geographical coordinates 55°61'86"N, 26°58'22"E). The background activity of ^{14}C was measured in the tree rings in Vaikšteniai village area (54°46'92"N, 24°77'81"E), located 165 km away from INPP in opposite to the predominant wind direction. That study revealed the local influence of the operating INPP. Moreover, the elevated ^{14}C activity levels were also measured in the tree rings corresponding to the period after the shutdown of Unit 1 on December 31, 2004 and Unit 2 on December 31, 2009. This observation was attributed to post-shutdown activities related to the dismantling of the plant equipment and management operations of radioactive waste at the INPP. Also, the precise measurement with the AMS provided evidence of the additional influence of the maintenance activities of the reactors at Ignalina NPP during the operational period of the plant.

In this article we provide a deeper insight into ^{14}C distribution in the tree rings in the close vicinity of Ignalina NPP. In this study, the pine trees have been investigated providing the net of the monitoring points around INPP. The main idea was to check whether the radiocarbon can be used as a natural indicator of the airborne releases during the NPP operation which is recorded as an activity pattern in the tree rings. Moreover, this is of particular importance for the past practices of NPP operations when ^{14}C releases were not registered and therefore the indirect measurement method like the tree-ring analysis serves as the unique possibility for the retrospective assessment.

Application of the highly sensitive ^{14}C measurement technique combined with environmental modeling can be used as a powerful tool for the NPP anthropogenic impact assessment.

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Table 1 Sampling point locations.

Sampling point	Distance from INPP (km)	Corresponding sector	Wind direction from INPP
N1	1.9	45°	NE
N2	6.6	22.5°	NNE
S1	1.8	180°	S
W1	2.6	270°	W
W2	4	270°	W

SAMPLING AND METHODS

Sample Collection

In order to evaluate the impact of Ignalina NPP (Lithuania) operation on the environment, 11 tree cores were collected in total. RBMK1500 reactor unit was designed with a 150 m high ventilation stack. Both INPP reactors used their own stack for efficient ventilation. The plume released at this height does not reach the ground and cannot be detected closer than a specific distance depending on atmospheric dispersion characteristics. Therefore, for optimal detection of the INPP releases, tree core samples were collected farther than about 1.5 km from the INPP stacks. The distance, where the maximal ground activity in the air is reached, was assessed by a Gaussian plume model assuming atmospheric stability class C.

Due to the limitation to the proximity of the NPP and possible sampling locations (availability of and accessibility to the pine trees), we were constrained to collect samples from strict distance band and positions around the NPP stacks.

Eight core samples were collected: 5 at the best available sampling sites around INPP (see Table 1), the other 3 samples from background site-village Vaikšteniai, located (54°46'92"N, 24°77'81"E) 165 km away from Ignalina NPP. The locations were recorded by GPS coordinates and are shown in Figure 1.

The majority of tree core samples (total 4) were taken at one site 1.9 km northeast of the INPP. This site was crucial to our study due to its geographic location and dominant downwind direction (geographical coordinates 55°61'86"N and 26°58'22"E). In our previous study, we observed a significant increase in atmospheric ^{14}C concentration in the peninsula of Drūkšiai lake during the period of Ignalina nuclear plant operation, from 1983 to 2009 (Ežerinskis et al. 2018). To evaluate distance effect in north-northeast direction, one more sample was collected at the site 6.6 km away from NPP (55°39'36"N, 26°35'50"E). The Other 3 locations were selected to cover different directions with different distances from NPP. Samples from west side from NPP where collected at 2.2 km (55°36'1"N, 26°31'38"E) and 4 km (55°36'16"N, 26°29'58"E) distances and one from south side 1.8 km (55°35'18"N, 26°33'50"E) away. A Background location at the village of Vaikšteniai was selected to avoid possible local anthropogenic atmospheric pollution from INPP, Vilnius city and other towns.

Each tree for sampling was selected using these requirements: species (*Pinus Sylvestris*), age >40 yr, no visible diseases, no physical damage to strain and branches, similar soil and humidity conditions to background area.

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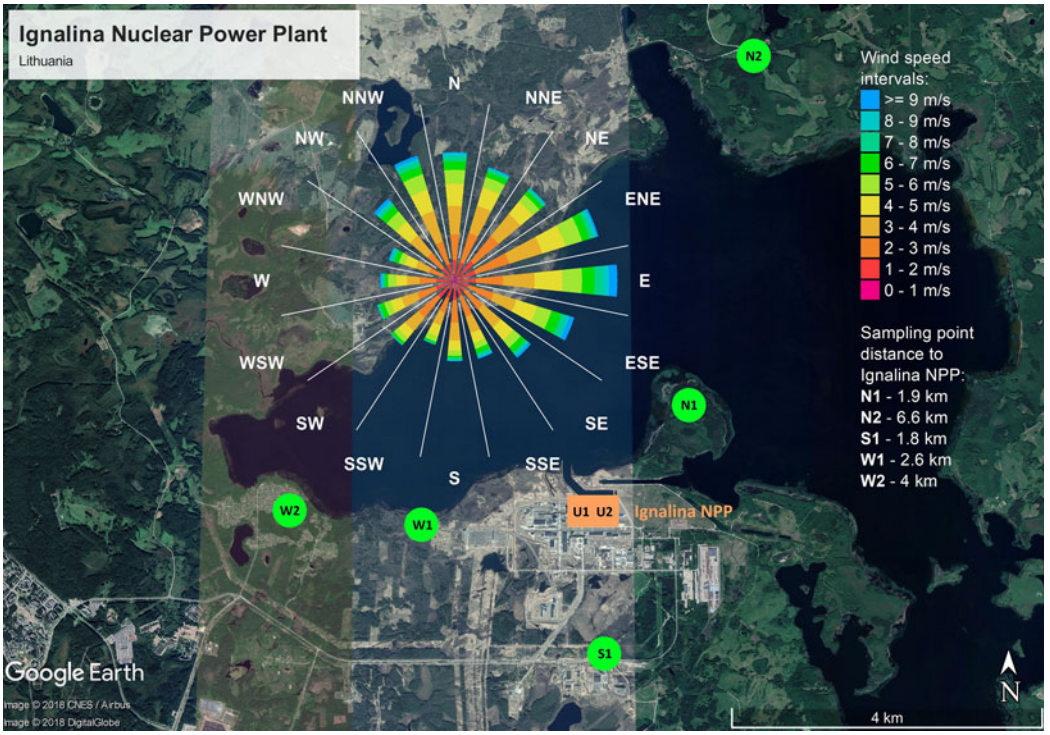

Figure 1 Location of the sampling sites around Ignalina NPP and dominant wind rose during warm and light (vegetation) period in 2004–2017 (wind direction off INPP).

Sample Preparation and ^{14}C Analysis

More than 400 wood and reference samples were prepared from the 11 tree cores collected in order to evaluate the impact of INPP operation on the environment. Our previous investigation (Ežerinskis et al. 2018) was extended by adding samples from 4 new locations and increasing sampling area to 5 spots. Trees, corresponding to above mentioned requirements, were selected and marked for further sampling. *Pinus Sylvestris* tree cores were extracted with 5 mm-diameter Swedish tree-core borer (Haglof®). From each tree, we extracted a minimum of 2 cores from different angles around tree trunk to reduce tree-ring counting errors and to get some insight into sample variety from the same tree. Furthermore, the most representative core sample used for further annual tree-ring preparation was selected from each tree. Moist and soft tree rings were separated and dried. Earlywood and Latewood from single tree ring were not separated.

The cellulose fraction was extracted performing BABAB (base-acid-base-acid-bleach) method (Němec et al. 2010). The end result of chemical pre-treatment is pure holocellulose.

Graphitization procedure was performed with AGE3 equipment (Ežerinskis et al. 2018, Wacker et al. 2010). Prepared graphitized samples were used to produce SSAMS targets for radiocarbon analysis. Typical SSAMS system parameters can be found in the paper Ežerinskis et al. (2018).

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The measurements are reported in units of pMC obtained by the *abc* program (version 7.0) from NEC. This ratio is constant over time accordant to this definition (Stuiver and Polach 1977; Donahue et al. 1990):

$$pMC = \frac{A_{SN}}{A_{ON}} \cdot 100\% \quad (1)$$

where A_{SN} is the specific activity of the sample, A_s, normalized to $\delta^{13}\,C = -25\%$; A_{ON} is the normalized activity of the standard.

Meteorological Observation Data

Dispersion of the atmospheric ^{14}C releases from NPP and availability for the assimilation by local plants is determined by meteorological conditions. Fortunately, local meteorological data was available from records of meteorological station at Ignalina NPP site for the period of 2004–2017. Meteorological readings were systematically taken at ground level and at the top of 40 m height meteorological tower. The data was recorded every 3 hr and it included: wind direction, average and maximum wind speeds, temperature, humidity, atmospheric pressure and the amount of precipitation. In our study data records were used to establish local wind roses and to determine vegetation periods for every year during 2004–2017. Correlations between wind blowing frequency towards sampling site and ^{14}C contamination in tree rings are examined further.

Local Scale ^{14}C Atmospheric Transport Modeling

During routine operation of INPP atmospheric discharges were released via the main ventilation stacks at the height of 150 m and are composed dominantly by radioisotopes of noble gases $^{85\,m,87}Kr$, ^{41}Ar, $^{133,135}Xe$, but also contain significant amounts of ^{3}H, ^{131}I, and ^{14}C. INPP operated two RBMK-1500 reactors: Unit 1, 1983–2004; Unit 2, 1987–2009. Since the very end of 2009 all INPP units have been in final shutdown condition and dismantling and decontamination activities of some less-contaminated equipment (turbine hall building equipment, emergency core cooling equipment, etc.) have been initiated.

The single circulation circuit design of RBMK leads to a continuous release from the coolant and discharge of ^{14}C through the stack mainly in the form of CO_2. A part of ^{14}C from the coolant is purified by ion-exchange resins or released as liquid discharges. Radiocarbon releases may also occur occasionally during reactor maintenance (e.g. due to changing of fuel channels [Ežerinskis et al. 2018]) or dismantling works.

The classical Gaussian transport model is suitable for the short-range dispersion modeling of the atmospheric discharges from the elevated source at steady state meteorological conditions. In case of varying weather during prolonged discharges, the model can still be applied if taking a superposition of sets of steady state events with averaged atmospheric conditions. The following Gaussian model equation determines the time-integrated atmospheric concentration of gas at any point in space, on the grid of x-axis, directed to the downwind from the release point, and y-axis in crosswind direction (according to Gifford 1960):

$$C(x,y,z,H) = \frac{Q}{2\pi\sigma_y(x)\sigma_z(x)u} exp\left[-\frac{1}{2}\left(\frac{y}{\sigma_y(x)}\right)^2\right]\left\{exp\left[-\frac{1}{2}\left(\frac{z-H}{\sigma_z(x)}\right)^2\right] + exp\left[-\frac{1}{2}\left(\frac{z+H}{\sigma_z(x)}\right)^2\right]\right\} \quad (2)$$

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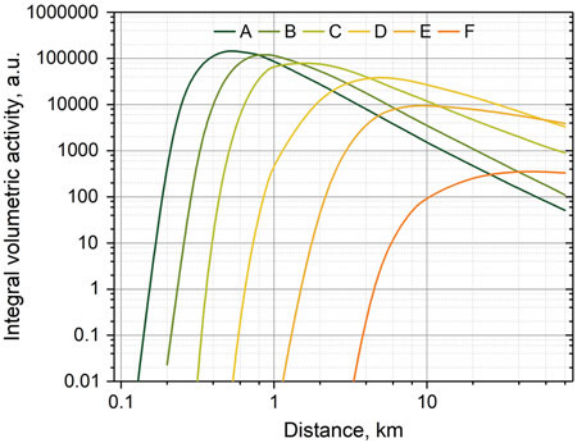

Figure 2 Distance dependent near ground integrated air activity assessed by Gaussian model for hypothetical gas amount releases from Ignalina NPP stack H=150 m for the cases of A to F atmosphere stability classes.

where: C is time-integrated atmospheric concentration (Bq s)/(m^3); Q is source term (Bq); H is effective release height H=150 (m); x is downwind distance (m); y is crosswind distance (m); z is vertical axis distance (m); σ_y is standard deviation of the integrated concentration distribution in the crosswind direction (m); σ_z is standard deviation of the integrated concentration distribution in the vertical direction (m); u is average wind speed at the effective release height (m/s).

Depletion and radioactive decay of ^{14}C is neglected because the near field around the source is considered (< 6.6 km). Briggs equations (Briggs 1973) are commonly used to determine σ_x and σ_y for A to F Pasquill atmosphere stability classes. Basing on (2) formula course of distance dependent near ground integrated air activity was calculated on plume centreline for some hypothetical amount of gas, released from the stack of Ignalina NPP at H=150 m, and it is presented in Figure 2 for different atmosphere stability classes. Calculations were performed by using HotSpot Version 3.0 software.

The actual amount and manner of ^{14}C releases from INPP (the source term in Bq) is not known for the whole studied period, because systematic measurements of radiocarbon discharges were implemented at INPP only from 2008. When operated, the maximal permissible level of ^{14}C release to atmosphere from INPP was set as $2.3{\cdot}10^{14}$ Bq/yr by the normative document (LAND 42 2007). Declared annual ^{14}C atmospheric discharges were about $1.3{\cdot}10^{11}$ Bq/yr (U1DP0 EIAR 2007) in the period of INPP two reactors operation before 2004. Since 2008 systematic measurements of the ^{14}C releases are available and the levels of ^{14}C annual discharges to the atmosphere are presented in the Table 2 (European Commission 2016).

Annual discharges ranging from $2.18{\cdot}10^{10}$ to $3.09{\cdot}10^{10}$ Bq/yr in the years of 2008–2009 when only the second reactor unit was in operation, and from $1.15{\cdot}10^{9}$ to $4.39{\cdot}10^{9}$ Bq/yr after INPP last reactor final shutdown at the end of 2009. Unfortunately, the time series of the ^{14}C release during the year was not provided.

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Table 2 Ignalina NPP declared annual discharges of ^{14}C to atmosphere (European Commission 2016).

	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017
Airborn ^{14}C releases from INPP, Bq/yrr	$2.18{\cdot}10^{10}$	$3.09{\cdot}10^{10}$	$1.42{\cdot}10^{9}$	$3.52{\cdot}10^{9}$	$2.07{\cdot}10^{9}$	$4.39{\cdot}10^{9}$	$2.19{\cdot}10^{9}$	$4.07{\cdot}10^{9}$	$1.25{\cdot}10^{9}$	$1.15{\cdot}10^{9}$

Once released from the stack, ^{14}C undergoes the process of transportation and dispersion in the atmosphere and then it is assimilated by the pine trees. The input data for the dispersion assessment was taken from the meteorological records database. Every 3 hr duration data record (i), corresponding to the warm period (T_{air} ~+5°C ÷ +35°C) and light time of the day (6 hr until 21 hr), was considered. Total number of suitable data records in 2004-2017 was N=15081. Space around NPP was divided into 16 directional sectors, having 22.5° angular width. All annual individual meteorological data records i of the certain year n light and warm period were grouped, corresponding to wind blowing to the certain sector j- $u_{i\to sector\_j}$. The annual (n) integrated activity at any distance x from INPP in the sector j at the near ground level z=1.5 m on the plume centreline y=0 m can be assessed by the following formula:

$$C_{sector_j,n}(x) = \frac{1}{2\pi\sigma_y(x)\sigma_z(x)} \sum_{i=1}^{N} \frac{Q_{i,n}}{u_{i\to sector_j,n}} \left\{ exp\left[-\frac{1}{2}\left(\frac{z-H}{\sigma_z(x)}\right)^2 \right] + exp\left[-\frac{1}{2}\left(\frac{z+H}{\sigma_z(x)}\right)^2 \right] \right\} \quad (3)$$

Prevailing atmosphere stability class was identified based on the ^{14}C tree-ring measurements taken in the same direction but at different distances from the source (N1, N2 and W1, W2 points). According to the Gaussian model, pattern of the activity change with the distance being unique for every class, therefore actual activity ratios N1/N2 and W1/W2 would correspond to the certain atmosphere stability class. Comparison of the calculated theoretical and measured activity ratios is presented in Table 3.

The results obtained for the N1/N2 and W1/W2 ^{14}C activity measurements gave median values of 3.79 and 0.92, correspondingly, which refer to an averaged dilution typical between C and D stability classes. Our choice was to admit prevailing stability class C and to use corresponding σ_x and σ_y values in the further assessment with the Gaussian model.

Actual course of the releases from the INPP stack $Q_{i,n}$ (every 3 hr during the year n) is not known, so, the only relative directional and distant dependent integral air activities $\widehat{C_{sector_j,n}}(x) = \frac{C_{sector_j,n}(x)}{\sum^{Q_{i,n}}}$, which is proportional to the dilution factor) in INPP vicinity can be obtained by Gaussian modeling for every year at every sampling site. Such relative annual measure allowed to eliminate influence of Q term, but preserved distance and direction specifics.

Further correlations between excessive ^{14}C measured in tree rings and normalized annual integrated air activity, assessed by using Gaussian model basing on meteorological data for 2004–2017 period of time, was studied for the sampling locations.

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Table 3 ^{14}C activity ratios for average atmosphere stability class identification from the tree-ring measurements and Gaussian model calculations.

Sampling locations	Theoretical air activity ratio for Pasquill stability classes						Median of measured tree-ring activity ratio
	A	B	C	D	E	F	
N1/N2	9.55	8.37	4.43	0.5	0.014	4.5E-09	3.79
W1/W2	2.26	2.08	1.74	0.78	0.23	0.0013	0.92

RESULTS AND DISCUSSION

The radiocarbon analyses provided critical data about radiocarbon releases and atmospheric transport from Ignalina NPP and gave the opportunity to estimate its environmental impact and to couple with the Gaussian model.

The results showed a pronounced increase of ^{14}C up to about 15 pMC (1999 y) in the tree rings during INPP operation period (Unit 1, 1983–2004; Unit 2, 1987–2009) as well as during decommissioning periods (see in Figure 3). We observe that from the beginning of INPP operation (1984–1994), the excess of ^{14}C was minor. An average excess of 3 pMC in this period was observed reaching a maximum value of 6 pMC. Year 1995 is a clear breaking point in INPP curve. From this point, the excess of ^{14}C almost doubles and lasts up to the shutdown of the second INPP reactor in 2009.

As expected, the effect of contamination reduction with distance was observed in average by 6 pMC by increasing distance from 2 to 6.6 km from the INPP. Directional and annual variations were also apparent.

Correlations between measured excess ^{14}C in tree rings and corresponding assessed annual radiocarbon relative integral air concentration at the sampling location were studied. The derived correlation coefficients for 2004–2015 are presented in the Table 4. Selected typical scatterplots for the several years of INPP exploitation (2004, 2007) and decommissioning (2009, 2012) periods are shown in the Figure 4.

A good correlation between measured and calculated ^{14}C activity was observed. The correlation coefficient R varied from 0.56 (2013) up to 0.98 (2005). This affirms, that the radiocarbon discharges from the Ignalina NPP atmosphere were done in a quite constant manner and can be well predicted by Gaussian model considering local wind rose. It also confirms, that ^{14}C measurements of the pine tree rings can be used as a tool of the retrospective monitoring of the radiocarbon discharges from the nuclear power plants.

Assessment of the Effective Dose to the Public

According to the data provided by the INPP and our measurement results, we can confirm that the INPP discharges ^{14}C into environment during its normal operations and also after the final shutdown of the both units due to the dismantling operations and management of radioactive waste. Thus, the additional effective dose to the local public can be estimated. We followed the effective dose due to ^{14}C assessment method using models that employ a specific activity approach which is proposed in IAEA publication Annex III (IAEA 2001). The models

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Table 4 Correlations between measured ^{14}C excess activity in tree rings and assessed by Gaussian model annual air integral activities $\widehat{C_{sector_j,n}}(x)$.

Year	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Correlation coefficient R	0.87	0.98	0.7	0.84	0.75	0.83	0.59	0.88	0.94	0.56	0.84	0.88

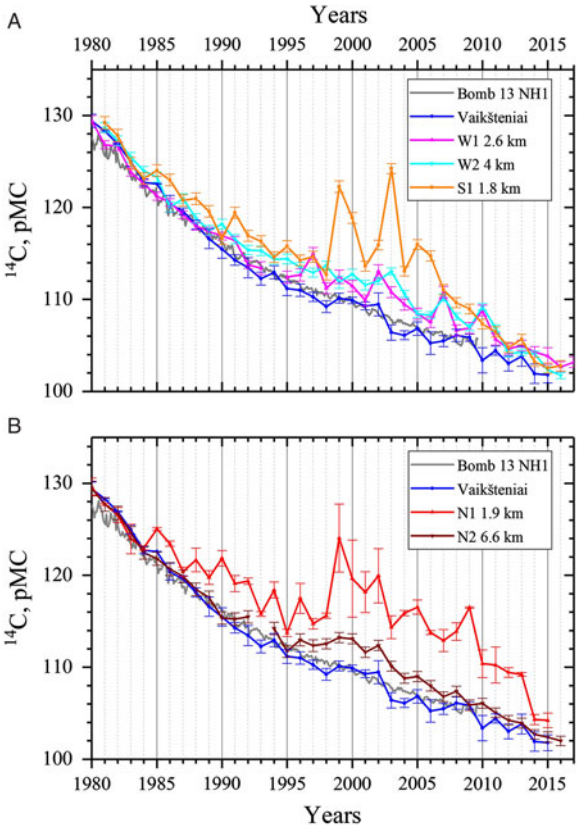

Figure 3 Trend of ^{14}C activity measurements in the tree rings from the background location in Vaikšteniai and sampling points around Ignalina NPP.

assume, that the ^{14}C released is associated with CO_2 molecules and is subsequently fixed within plant tissues during photosynthesis and are transported along with stable ^{12}C through food chains into the human body. Ingestion of carbon originating from the atmosphere is the primary mode of exposure (all other pathways of exposure will contribute less than 1% of the total dose). This method provides a conservative dose estimate, because an individual is assumed to be in complete equilibrium with maximum levels of environmental specific activity of ^{14}C and the ratio between the radionuclide and its stable counterpart is fixed. According to the method, the dose rate at equilibrium will be directly proportional to the concentration of ^{14}C in air relative to the concentration of stable carbon at a given location

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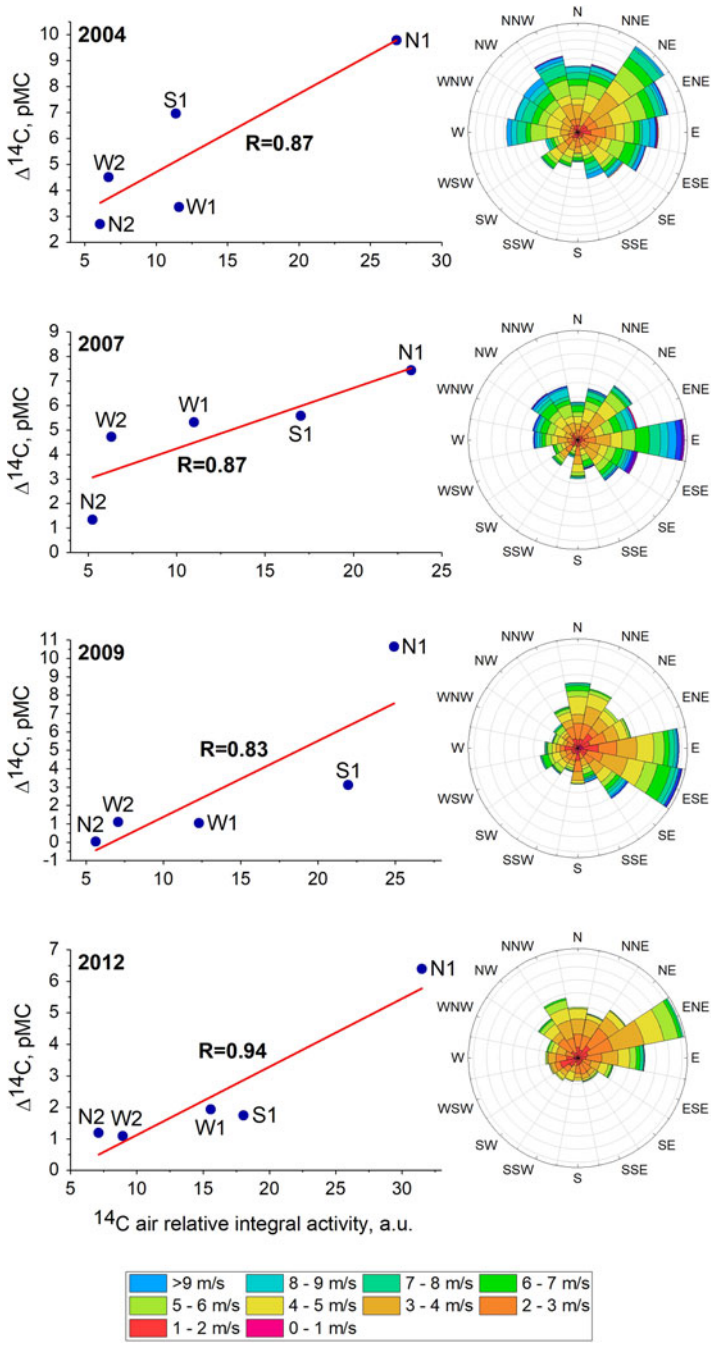

Figure 4 Typical scatterplots between measured ^{14}C excess activity in tree rings and assessed by Gaussian model annual air integral activities Csectorj, nx for the sampling locations around Ignalina NPP.

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x, multiplied by the fraction of total dietary carbon derived from this location (IAEA 2001):

$$D^{max} = (A)_x^{max} \cdot (F_{c,a})_x \cdot g \quad (4)$$

where: D^{max} is the effective dose rate (Sv/a); $(A)_x^{max}$ is the maximum specific activity (Bq ^{14}C/g C) to which food products at location x will be chronically exposed; $(F_{c,a})_x$ is the fraction of total dietary carbon derived from location x by the representative member of the critical group (conservatively assumed to be unity); g is the effective dose rate factor that relates the annual dose rate (Sv/a) to the concentration of ^{14}C per gram of carbon in people (Bq/g) (recommended value for screening is $5.6{\cdot}10^{-5}$ Sv/a per Bq/g C (IAEA 2001).

Values of specific activity $(A)_x^{max}$ in Bq ^{14}C per gram of C can be derived from the tree-ring ^{14}C measurements in pMC by using the relationship of 1 pMC as equivalent to 0.00226 Bq/g C (Stuiver and Polach, 1977). This would provide an effective dose estimate of about 0.12 μSv/yr per 1 pMC. This value is similar to conservative estimate of 0.18 μSv/yr per ^{14}C excess by 1 pMC (Mažeika et al. 2008), obtained by combining ^{14}C release rate from INPP assessment by Gaussian plume model backward calculations with the nuclide-specific atmospheric routine release dose conversion factor, representing the ratio of the annual effective dose for a farmer critical group member (Sv/yr) at the location x of the highest predicted radionuclide concentration in air, to the activity released from the INPP 150 m height stack (Bq/yr) (Motiejūnas et al. 1999; LAND 42 2007).

The effective dose rate caused by INPP discharges of ^{14}C were estimated to be for 1984-1993: 0.4 μSv/yr; 1994-2009: 0.8 μSv/yr; 2009-2015: 0.6 μSv/yr. An average effective dose rate for the time period of 1984-2015 is equal to 0.6 μSv/yr. The maximum value of 1.8 μSv/yr was reached in 1999. These effects were caused by INPP repair works, mostly by replacing nuclear reactor core channel tubes. The core modernization started in 1998, then 3 reactor channel tubes were replaced. In 1999, 100 channel tubes; in 2000, 7; in 2001, 50; in 2002, 75; and in 2003, 10 (Ežerinskis et al. 2018). Later on, INPP dismantling activities contributed significantly to ^{14}C releases. Therefore, in time span 1995–2009, we have observed elevated ^{14}C activities in tree rings with a maximum of 15 pMC and an average value of 7 pMC.

Monitoring results acquired by Xu et al. (2016) showed a maximum of 15.8 pMC increases in radiocarbon activity near the Fukushima boiling water reactors in tree rings from 2000, with estimated additional effective dose of 2.0 μSv/yr.

Concerning ^{14}C releases from other type reactors (PWR), Roussel-Debet et al. (2006) reported results from 14 sampling sites around French NPPs. Results from those measurements shows that discharges made a very slight effect on ^{14}C activity, Δ^{14}C varying from 101‰ (110.1 pMC) in the non-influenced zone to 123‰ (112.3 pMC) in the influenced zone. This increase in dose is negligible and contributes less than 0.1 μSv/yr.

Janovics et al. (2017) performed research to determine the effects of Paks NPP (PWR type reactor of VVER design) operation to environment before and after a third-level incident (INES-3). Tree samples measurements did not show a noticeable increase in ^{14}C background. This could be due to the short-term manner of accidental release and dominant routine releases of ^{14}C from PWR in organic form which is not assimilated by plants. Besides, sampling sites were located to close (250 m and 400 m) to Paks NPP for elevated plume (130 m) to reach ground with the highest concentration.

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Furthermore, atmospheric radiocarbon has been monitored around Jaslovské Bohunice NPP (VVER) and relatively higher excess exposure dose of 3 μSv/yr to local public was evaluated (Povinec et al. 2008). A higher value was obtained, presumably, as a result of extracting ^{14}C depleted background level, which was considerably affected by use of fossil fuels. In any case, the resulting dose was negligibly small as to compare with the natural exposure background and demonstrated that radiocarbon releases from NPPs did not pose radiological hazard to the public.

CONCLUSIONS

This study reports the annual ^{14}C variations in Lithuanian pine tree rings from 1957–2015. The Ignalina nuclear power plant (Ignalina NPP) operation time period was studied in more detail. Results demonstrate that the first 12 yr (1983–1995) during normal INPP operation the excess of ^{14}C was negligible with an average value of 3 pMC. However, the excess of ^{14}C during the time span (1995–2009) almost doubles to maximum of 15 pMC (the average value enhances to 7 pMC) and the absence of the correlation with the background in this period lets us assume that the additional ^{14}C contamination source makes a significant contribution to the total ^{14}C discharges from Ignalina NPP.

The estimated effective dose rate caused by the INPP operations for the time period 1984–2009 on average was 0.6 μSv/yr, which is negligible compared to the natural background dose of 2.5 mSv/yr.

Distance correlation analysis shows a significant decrease of ^{14}C concentration in farther distances from NPP units highly corresponding to wind direction. Implementing wind speed and directional data from different heights provides the necessary information for ^{14}C distribution and dissipation model around NPP. Gaussian ^{14}C dispersion model reconstructs how radiocarbon was released from NPP in definite time span, recreating historical events of Ignalina NPP. The analysis of wind rose data and ^{14}C concentrations around Ignalina NPP shows a high correlation, indicating greater flora sensitivity for ^{14}C emissions and model eligibility.

The results of correlation analysis of wind direction and ^{14}C concentration in the tree rings provide confirmation that ^{14}C measurements in the tree rings can be used as a tool of indirect assessment of gaseous discharges from NPP during its operation as well as post-operational periods. The data of our study could be used for estimations of future ^{14}C release from NPP as well as for the modeling of environmental impact of ^{14}C on inhabitants, fauna, and flora.

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