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Carbon-14 in tree rings and other terrestrial samples in the vicinity of Ignalina Nuclear Power Plant, Lithuania

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Abstract

The results of ^{14}C measurements in the annual tree rings from the Ignalina Nuclear Power Plant (INPP) surroundings, Lithuania, for the period of its operation from 1984 to 2002 are presented. The terrestrial samples, mainly moss and related soil, are studied in places as well. The tree rings have shown slightly enhanced ^{14}C activity due to operation of the nuclear power plant. The maximal calculated normalized ^{14}C release of 11 TBq GW_e^{-1} $year^{-1}$ and the maximal effective dose of 2.0×10^{-3} mSv $year^{-1}$ resulting from the ^{14}C were estimated for 1999. For other years of INPP operation these values are lower. The excess of ^{14}C specific activity measured in the moss and soil samples from moss-covered sites near the nuclear power plant (up to 0.5 km) showed highly elevated ^{14}C contents (up to 813 pMC), probably indicating releases of particulate material.

Keywords: Ignalina NPP; Carbon-14; Tree rings; Airborne release; Environment; Dose

1. Introduction

Carbon-14 (^{14}C, half-life 5730 ± 40 year, maximum beta energy of 156 keV) occurs naturally in the environment due to the cosmic-ray induced production in the atmosphere (Libby, 1946; Raaen et al., 1968). At the beginning of the 19th century, the concentration of ^{14}C, compared to the stable isotope ^{12}C, in the atmosphere began to decrease due to the increased burning of fossil fuel (Fairhall and Young, 1970). In the 1950s, when many atmospheric nuclear weapon tests took place, the ^{14}C concentration in air rose sharply to a maximum (double the natural concentration), and has gradually decreased after 1960 (Hertelendi and Csongor, 1982; Levin and Kromer, 2004).

Carbon-14 is also artificially produced in all types of nuclear reactors, including nuclear power plants (NPPs), mainly through neutron-induced reactions with isotopes of carbon, nitrogen and oxygen present in the fuel, cladding, coolant, moderator and structural materials of the reactor (NCRP, 1985; IAEA, 2004). A fraction of the generated ^{14}C is released continuously during normal operation of NPPs, mainly in two chemical forms; oxidized, i.e. carbon dioxide (CO_2), and reduced, which mostly is in the form of CH_4 (Levin et al., 1988). Because of the easy incorporation of

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^{14}C in CO_2 form into the global carbon cycle, rapid mobility, long ^{14}C half-life and particularly, the considerable inventory or high release rate (average-normalized release rates from 1990 to 1994 in TBq GW_e^{-1} $year^{-1}$ in brackets for respective nuclear power reactors: CANDU (1.6) > RBMK (1.3) > BWR (0.51) > PWR (0.22) (UNSCEAR, 2000), these releases should be considered very carefully in estimating their local and global effect on the individual effective dose and collective dose commitments (McCartney et al., 1986; Aquilonius and Hallberg, 2005).

For all types of reactors, except pressure water reactors (PWRs), most of the gaseous releases of ^{14}C are in the form of $^{14}CO_2$ (IAEA, 2004), which is easily assimilated by plants through photosynthesis. As a consequence, increased specific activity of ^{14}C may be found in surrounding plants and all other types of biota.

In case of the Ignalina NPP (INPP), Lithuania, the importance of ^{14}C is also due to the large total mass of graphite per reactor of more than 1800 tons, and the consequently there is a large ^{14}C inventory in graphite (7.0×10^{14} Bq in all the irradiated graphite constructions of a reactor) (Ancius et al., 2005; Maceika et al., 2005). It is the upper limit estimate, derived assuming that activation products are retained in the graphite.

Apart from ^{14}C in CO_2 and hydrocarbon forms, the contribution of ^{14}C present in particulates from airborne releases is also considered to be significant for certain types of nuclear reactors – advanced gas-cooled reactors (AGRs), channelled large power reactors (Russian acronym – RBMKs) (Bush et al., 1984; Marsden et al., 2002).

Measurements of the ^{14}C specific activity in environmental samples are therefore useful for estimating the doses to public through terrestrial food chains. Among the variety of environmental samples, tree rings represent the atmospheric $^{14}CO_2$ concentration of respective vegetation year due to steady-state of carbon containing reservoirs – atmospheric CO_2 and terrestrial plants (wood cellulose). Tree rings were used widely to reconstruct atmospheric $^{14}CO_2$ variations in the past (Tans et al., 1979).

According to Lithuanian regulation (LAND 42–2001), the Ignalina NPP is required to assess doses due to releases of radionuclides during normal operation. Considering ^{14}C, it is stated in LAND 42–2001 that ''in the case of airborne and aquatic discharges, ^{14}C activity shall be measured or estimated and validated for various operation conditions''. Taking into account this requirement, the Ignalina NPP is going to implement a ^{14}C-in-airborne-releases monitoring program with continuous air sampling in a bypass of the main exhaust air stream in the stack by a differential CO_2/CH_4 sampler (Hertelendi et al., 1989).

Since the beginning of the Ignalina NNP operation, the presence of ^{14}C in the environment has been monitored in tree rings, annual and perennial aquatic and terrestrial plants, surface water and (on a smaller scale) groundwater, and other environmental samples (Mazeika, 2002). The annual terrestrial plants were investigated in the greatest detail. For example, the specific activity of ^{14}C ranged from 96 to 123 pMC (pMC means percent of modern carbon, 1 pMC = 2.27 Bq kg^{-1} C) (Stuiver and Polach, 1977; Stuiver, 1983) in wormwood (1996), and from 107 to 178 pMC in alder leaves (2001). In both cases, the highest specific activity of ^{14}C in wormwood (123 pMC) and alder leaves (178 pMC) was detected to the northeast of the Ignalina NPP. This direction coincides with the dominant wind direction and the zone of maximum impact of releases from the NPP.

This study includes the measurements of ^{14}C in tree rings representing the period of 1951–1995 for background Prienai trees, the period of 1983–2002 for background Varena trees and the period of NPP operation from 1984 to 2002 for Ignalina NPP trees, and terrestrial samples, mainly moss and related topsoil, of Ignalina NPP vicinity. The aim of the present investigation was to study a temporal and spatial distribution of ^{14}C specific activity in environmental samples in order to indirectly derive the magnitude of the annual release rate of ^{14}C and consequently, to assess maximal doses for the critical group members.

2. Site and environmental settings

The Ignalina NPP is located in the northeastern part of Lithuania near the borders of Belarus and Latvia (Fig. 1). The plant consists of two RBMK–1500 reactors. The ''1500'' refers to the designed electrical power in units of MW. Its designed thermal rating is 4800 MW. The nominal thermal power is 4250 MW, and the nominal electrical power is 1300 MW. The RBMK is a graphite-moderated boiling water, channel-type reactor (Almenas et al., 1998). This means that each fuel assembly is located in separately-cooled fuel channels, all surrounded by graphite. The two reactor units, Unit 1 and 2, were put into operation in December 1983 and August 1987, respectively.

As a part of the obligations of the European Union Accession Treaty, Lithuania is required to shut down Units 1 and 2 of the INPP and to decommission them as soon as possible. Unit 1 was shut down on December 31, 2004 and Unit 2 is to be shut down by 2010. In 2002, the Government of Lithuania approved the adoption of the ''immediate

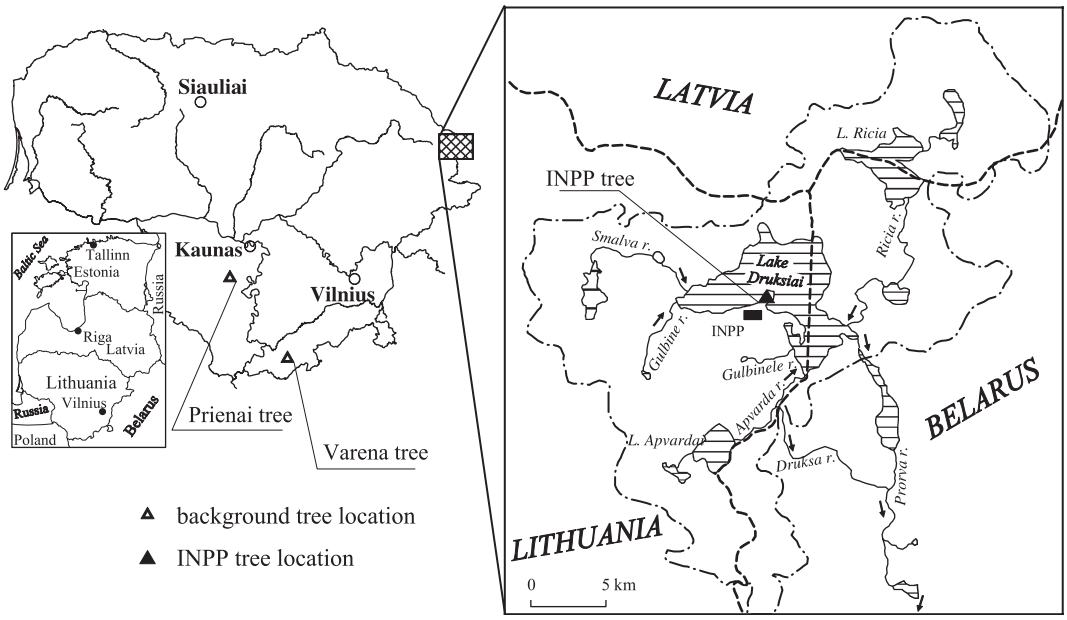

Fig. 1. Location of the trees used for a tree-ring sample preparation. More precise location of the tree near the Ignalina Nuclear Power Plant (INPP) is shown on the Lake Druksiai catchment sketch map.

dismantling'' strategy for decommissioning of both these units. A key component of this decommissioning strategy is to dispose of operating and decommissioning waste in a near-surface repository (NSR) that should become operational by 2011. This NSR should have a capacity for disposal of approximately 100 000 m^3 of low- and intermediate-level, short-lived radioactive waste. However, the option for irradiated graphite disposal is not decided yet.

The plant uses Lake Druksiai as a natural reservoir for cooling water (Fig. 1). The surface area and volume of the lake are 4.9×10^7 m^2 and 3.7×10^8 m^3, respectively. The maximum depth is 33 m, while the average depth is 7.6 m. Lake Druksiai is a flow-through lake with six small creeks flowing in and one river, with a water regulation dam, flowing out. The cooling water inlet and heated water outlet channels and industrial rainwater drains are shown in Fig. 2.

The routine airborne releases are discharged via the main stacks of 150 m height. Releases of particles escaping during the treatment of the radioactive waste are important too and for certain years, this pathway releases much more radionuclide than the main stacks to the environment (Motiejunas et al., 1999). The equipment for processing of radioactive waste coming from the both units is located in the western part of the industrial site and has separate ventilation systems with stacks of 75 and 12–13 m heights. The dominating wind direction is from the west or southwest. About 200 000 inhabitants live within a radius of 30 km of the power plant. The largest cities are: Daugavpils in Latvia with the population of about 126 000 (30 km away) and Visaginas with the population of about 28 800 (6 km away). The latter is the place of residence of the INPP employees.

3. Methods of investigation

3.1. Sampling (tree, moss and related soil)

The samples included tree rings, as well as moss and soil (up to 1 cm) from the layer beneath the moss vegetation collected at a short distance (0.5–2 km) from the power plant.

We have selected a pine tree in an area located in the vicinity of the INPP (geographical coordinates 55°36′49″ N and 26°34′53″ E, elevation 156 m a.s.l., 1.4 km to northeast from the stack of Unit 2) in a wet pine forest on 8 June 2003. Several cross-sections of the trunk of the model tree were taken for separating of annual tree rings. In order to analyse regularities of annual growth, besides the model tree, 10 more pine trees were sampled by an increment corer taking two cores per tree. The annual ring width fluctuations

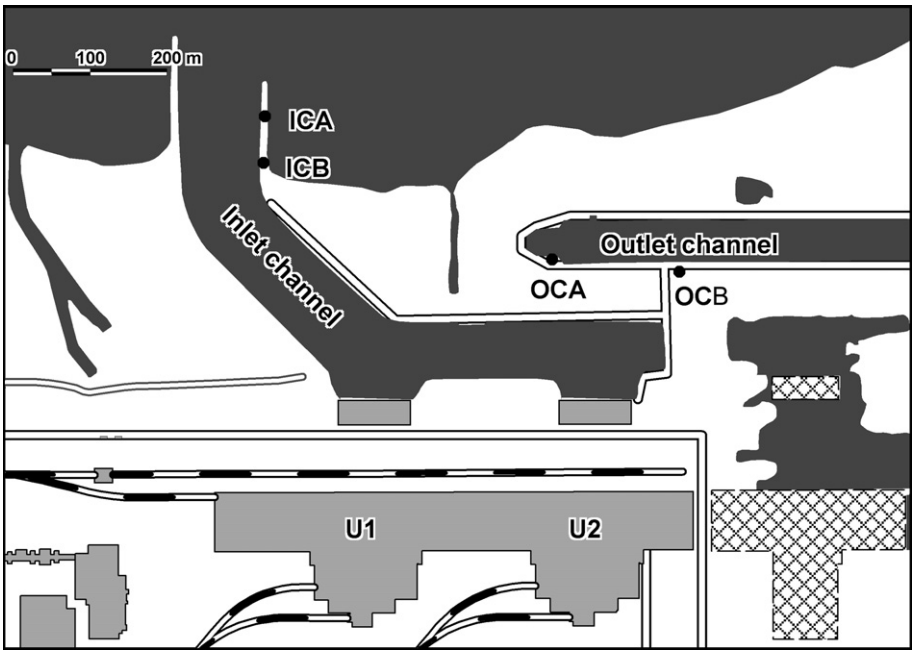

Fig. 2. Sites of collection of the moss and soil samples within the 500 m area to the north from the Ignalina NPP on 21 June 2005. U1 and U2 stand for Unit 1 and Unit 2. Label ICA means cooling water inlet channel, point A; ICB – cooling water inlet channel, point B; OCA – heated water outlet channel, point A; OCB – heated water outlet channel, point B. The pipe system from the reactor units to outlet channel is built underground.

of the model tree were highly synchronous with those of the other trees, and this supported the tree ring scale. Synchronic growth fluctuations indicate a common growth reaction to environmental factors and characterize the model tree as a good representative of the stand. This area was considered as mostly affected by airborne release from the INPP.

The pine tree from the background area (Prienai) was already analysed in previous studies (Mazeika, 2002). Another pine tree was selected from a dry pine forest in Varena district (geographical coordinates 54°02′30″ N and 24°42′30″ E, 153 m a.s.l.), that is 210 km to the southwest from the INPP, on 23 August 2002. In addition to the model tree, we have sampled 12 more pines using an increment corer, taking two cores per tree for annual growth analysis. For the Varena tree the ^{14}C results were obtained only for short period (1983–2002), corresponding to period of INPP operation.

Moss and soil sampling was carried out on 21 June 2005. The sampling locations are shown in Figs. 1 and 2.

All samples were dried in an oven (at 45 °C). After drying, any foreign material was manually removed from the moss samples. The dried soil samples were sieved through a 2-mm mesh and any roots and plant material were manually removed. The annual tree rings and moss samples were pyrolised (in airless conditions) to carbon without any chemical pre-treatment in order to convert bulk organics into a counting form. Soil samples were oxidized with MnO_2 in a reactor tube.

3.2. Analytical procedure

The specific activity of ^{14}C in environmental samples (tree rings, moss and soil) was measured (as described in Gupta and Polach (1985), Arslanov (1985) and Bowman (1995)). A conventional method for synthesis of benzene was applied (Kovaliukh and Skripkin, 1994). The main following procedures were followed: (1) reaction of carbon containing sample material with Li in the metallic reactor at the temperature 550–600 °C receiving Li_2C_2, (2) hydrolysis of Li_2C_2, receiving C_2H_2, (3) synthesis of benzene by cyclotrimerization of C_2H_2 using a catalyst containing aluminous-silicate and activated with V_2O_5, (4) purification of benzene from moisture remains by adding metallic sodium and distillation, and (5) adding the scintillation admixtures, PPO and POPOP, to benzene sample. The ^{14}C specific activity of benzene was determined by liquid scintillation counting. The main performance parameters of the spectrometric system for ^{14}C in benzene form with 3 mL Teflon vials were as follows: background count rate of 0.41 ± 0.04 CPM; counting efficiency of $71.3 \pm 0.8\%$; and Figure of Merit (Efficiency^2/Background) of $12\,380 \pm 700$.

The ^{14}C results were presented as specific activity. The uncertainty of the results (statistical radiometric error) was reported at 2 and 1 sigma-level for the tree rings samples and moss (related soil) samples, respectively. The quality of ^{14}C determinations in the

Radioisotope Research Laboratory was periodically tested through participating in various inter-comparisons. The results of Fourth International Radiocarbon Inter-comparison (FIRI), organized by the University of Glasgow in 2000, indicated that most of the ^{14}C results are of normal (3%) precision, with some of high (1.5%) precision.

4. Results and discussion

The results of the ^{14}C analysis in annual tree rings from the INPP area and from background regions (Prienai and Varena), together with the ^{14}C in air of the Northern Hemisphere data (Nydal and Lövseth, 1983, 1996), are presented in Table 1 and Figs. 3 and 4.

The highest specific activity of ^{14}C (199 pMC) was determined in an annual tree ring of the Prienai tree (background area) in 1964. According to the data of the Varena tree, the specific activity of ^{14}C has been reduced 1.8 times, with small variations in later years, and reached 110 pMC in 2002. In order to find out the ^{14}C excess in the annual rings of the INPP tree during the operation period of 1984–2002, the mean values of specific activity of ^{14}C in the rings of Prienai and Varena trees were used as the background specific activity of ^{14}C for 1983–1995; background values were estimated from the data of the Varena tree for 1996–2002. A comparison of the two data sets over time span 1983–1995 shows their coincidence within error limits (2σ) (Fig. 3). The background ^{14}C data for the whole investigation period are in a good correlation (correlation coefficient between two data sets is 0.996) with the ^{14}C in air of the Northern Hemisphere (Nydal and Lövseth, 1983, 1996) (Fig. 3).

A more detailed comparison of the background ^{14}C data with the ^{14}C data of the INPP tree for almost the whole operation period revealed a ^{14}C excess in tree rings near the power plant (Table 1, Fig. 4). At the beginning of the INPP operation, the ^{14}C excess was negligible whereas in 1996–2002, the ^{14}C excess value was the highest, reaching up to 5–11 pMC.

Based on the experimentally-determined values of the ^{14}C excess in the Ignalina NPP tree rings, an inverse problem was analysed – estimating ^{14}C release data based on the ^{14}C specific activity in the environment. The following parameters, important from the radiation and environmental protection point of view, were roughly estimated: air concentration of ^{14}C, as predicted by airborne releases; annually released activity of ^{14}C; normalized to electrical power release rate of ^{14}C; and annual effective doses to critical group, resulting from ^{14}C airborne release.

The main assumptions of these estimates are based on routine release conversion factors for the farmer (gardener) exposure pathway provided in Motiejunas et al. (1999) and Nedveckaite et al. (2000). Following the international recommendations (IAEA, 1998), a revised Ignalina NPP limitation system for the atmospheric and aquatic pathways was implemented (Nedveckaite et al., 2000; LAND 42–2001). In the case of an atmospheric release, the nuclide-specific routine release conversion factor represents the ratio of the annual effective dose for a critical group member (Sv year^{-1}) at the location of the highest predicted radionuclide concentration in air, to the activity released from the Ignalina NPP (Bq year^{-1}). The atmospheric dispersion model used in Motiejunas et al. (1999) and Nedveckaite et al. (2000) was the straight-line trajectory Gaussian plume model. For example, the dose conversion factor for the ^{14}C released from the 150-m high stack was 1.8×10^{-19} Sv Bq^{-1}. A radionuclide transfer factor in air representing the ratio of the annual mean air concentration (Bq m^{-3}) to the released activity (Bq s^{-1}) was used for the estimation of the ^{14}C concentration in air. The ^{14}C transfer factor in air of 1.75×10^{-8} s m^{-3} was estimated for the maximum affected zone in the INPP environment. We assumed that investigated INPP tree was located in this zone.

The carbon dioxide (CO_2) concentration in the atmosphere has increased due to human activities from 280 ppmv at the beginning of the industrial revolution to over 370 ppmv at present (Leuenberger et al., 1992; Keeling et al., 1995). Assuming an equilibrium between the $^{14}CO_2$ in air and ^{14}C in green plants, where the CO_2 concentration in atmosphere is equal to 330 ppmv, the ^{14}C excess in biota in terms of pMC can be linked to ^{14}C concentration in air as follows: 1 pmC equal to 0.00055 Bq m^{-3} (Vokal and Kobal, 1997).

Based on the above-mentioned assumptions and the transfer factors determined in Nedveckaite et al. (2000), the main calculation results are given in Table 2.

During 1984–2002, the excess ^{14}C concentration in air ranged from 5.5×10^{-4} to 6.1×10^{-3} Bq m^{-3}, the annually released activity of ^{14}C from 10^{12} to 10^{13} Bq, the normalized release rate of ^{14}C from 0.7 to 11 TBq GW$_e^{-1}$ year^{-1}, and the annual effective dose from 1.8×10^{-4} to 2.0×10^{-3} mSv year^{-1}, respectively. The minimal indefinite values of these parameters were identified in 1988 and 1993 and the maximal values in 1999, respectively.

The release of ^{14}C based on the given estimation is the highest among the releases of other radionuclides from the Ignalina NPP and contributes the most to the doses (Motiejunas et al., 1999). Therefore, the issue of ^{14}C releases from

Table 1
The ^{14}C specific activity (in pMC; 1 pMC = 2.27 Bq kg^{-1} C) in the tree rings

Year	Prienai tree	Varena tree	INPP tree	Background	^{14}C excess
	pMC ± 2σ	pMC ± 2σ	pMC ± 2σ	pMC ± 2σ	pMC ± 2σ
1951	101 ± 3				
1952	102 ± 2				
1953	102 ± 2				
1954	100 ± 2				
1955	102 ± 2				
1956	107 ± 2				
1957	111 ± 2				
1958	119 ± 3				
1959	125 ± 3				
1960	125 ± 2				
1961	124 ± 2				
1962	140 ± 2				
1963	189 ± 4				
1964	200 ± 6				
1965	178 ± 3				
1966	175 ± 4				
1967	167 ± 8				
1968	169 ± 7				
1969	162 ± 6				
1970	160 ± 4				
1971	155 ± 5				
1972	151 ± 6				
1973	146 ± 2				
1974	143 ± 2				
1975	142 ± 2				
1976	136 ± 3				
1977	139 ± 5				
1978	134 ± 2				
1979	132 ± 2				
1980	130 ± 2				
1981	130 ± 2				
1982	127 ± 2				
1983[a]	125 ± 2	123 ± 2	124 ± 3	124 ± 3	–
1984	121 ± 2	125 ± 2	124 ± 3	123 ± 3	1 ± 4
1985	122 ± 2	123 ± 2	124 ± 6	123 ± 2	1 ± 5
1986	120 ± 1	120 ± 2	121 ± 3	120 ± 2	1 ± 4
1987[b]	119 ± 1	117 ± 3	123 ± 4	118 ± 3	5 ± 5
1988	117 ± 1	120 ± 1	119 ± 4	119 ± 2	<1
1989	117 ± 1	115 ± 1	119 ± 3	116 ± 2	3 ± 4
1990	115 ± 1	117 ± 1	121 ± 3	116 ± 2	5 ± 4
1991	116 ± 1	114 ± 2	118 ± 3	115 ± 3	3 ± 4
1992	115 ± 1	116 ± 1	117 ± 3	116 ± 1	1 ± 3
1993	113 ± 1	112 ± 1	113 ± 3	113 ± 2	<1
1994	114 ± 1	113 ± 2	117 ± 2	114 ± 3	3 ± 4
1995	114 ± 1	108 ± 2	112 ± 3	111 ± 3	1 ± 4
1996		110 ± 1	117 ± 2	110 ± 1	7 ± 2
1997		110 ± 1	119 ± 2	110 ± 1	9 ± 2
1998		111 ± 1	116 ± 3	111 ± 1	5 ± 3
1999		110 ± 2	121 ± 1	110 ± 2	11 ± 2
2000		110 ± 2	117 ± 1	110 ± 2	7 ± 2
2001		108 ± 2	117 ± 1	108 ± 1	9 ± 2
2002		110 ± 1	119 ± 1	110 ± 1	9 ± 2

Values of ^{14}C specific activity in the annual rings of the Prienai tree for 1951–1982, mean values of ^{14}C specific activity in the annual rings of the Prienai and Varena trees for 1983–1995 and in the annual rings of the Varena tree for 1996–2002 given as background values. Only statistical radiometric uncertainty (2σ) is reported.

[a] Unit 1 put into operation in December 1983.

[b] Unit 2 put into operation in August 1987.

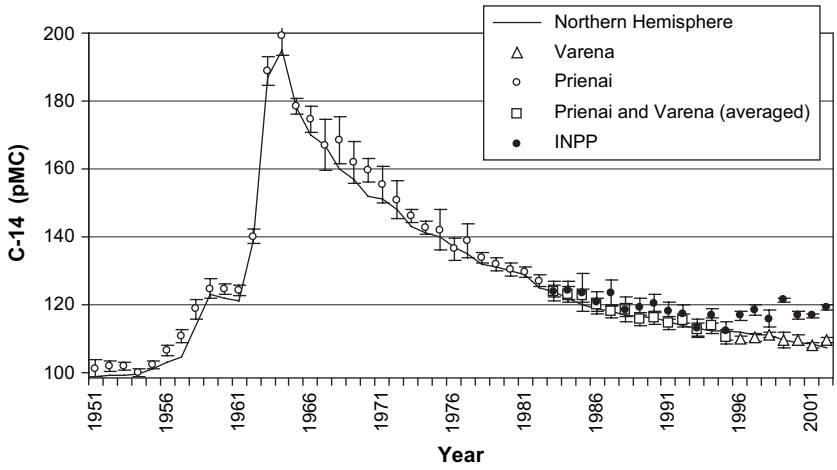

Fig. 3. The ^{14}C specific activity of pine tree rings in Lithuania, 1951–2002, compared with ^{14}C activity in air of Northern Hemisphere according to Nydal and Lövseth (1983, 1996).

the Ignalina NPP and their distribution pattern in the environment are of great importance and require further assessment.

The analysis of annual plants (tree leaves and grass) in the Ignalina NPP area showed, in some cases, even higher ^{14}C excess than that in the tree rings. The highest concentration of ^{14}C observed in seasonal vegetation samples (tree leaves and grass) in 2001–2003 was 33 pMC higher than the contemporary background level (106 pMC) measured in rushes (Magnusson et al., 2007). In 2001, the highest specific activity of ^{14}C in the plants of the Ignalina NPP environment reached 178 pMC. After the subtraction from this value of the background value (110 pMC), it turned out that the ^{14}C excess of about 68 pMC in annual plants was caused by the releases from the Ignalina NPP (Jakimaviciute-Maseliene et al., 2003). Unlike the annual tree rings, the annual plants may receive additional ^{14}C from a particulate material. This is especially characteristic of the moss and topsoil samples, which were briefly touched in this study (Table 3).

Only five moss samples (dry mass – up to 30 g per sample) and two samples of soil with humus (dry mass – 80 g per sample) just below moss were measured from the vicinity of INPP. However, moss and especially soil samples showed very wide range of ^{14}C excess, from 8 to 457 pMC, and from 96 to 706 pMC, respectively.

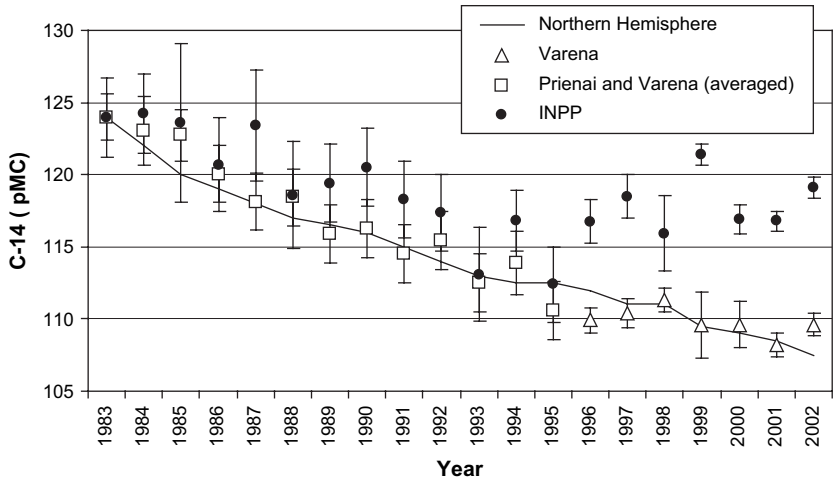

Fig. 4. The ^{14}C specific activity of pine tree rings in Lithuania for the period of Ignalina NPP operation 1984–2002, compared with ^{14}C activity in air of Northern Hemisphere according to Nydal and Lövseth (1983, 1996).

Table 2
The ^{14}C excess based on the tree rings measurements, respective excess air concentrations of ^{14}C, calculated stack release during one year and annual doses for the Ignalina NPP operation period 1984–2002

Year	^{14}C excess (pMC)	Computed air concentrations of ^{14}C (Bq m^{-3})	Calculated release during one year (Bq year^{-1})	Normalized release rate (TBq GW$_e^{-1}$ year^{-1})	Annual effective dose to adult gardeners resulting from ^{14}C airborne release (mSv year^{-1})
1983[a]	–	–	–	–	–
1984	1	5.5×10^{-4}	1.0×10^{12}	1.9	1.8×10^{-4}
1985	1	5.5×10^{-4}	1.0×10^{12}	1.0	1.8×10^{-4}
1986	1	5.5×10^{-4}	1.0×10^{12}	1.0	1.8×10^{-4}
1987[b]	5	2.8×10^{-3}	5.0×10^{12}	4.8	9.0×10^{-4}
1988	<1	$<5.5 \times 10^{-4}$	$<1.0 \times 10^{12}$	<0.7	$<1.8 \times 10^{-4}$
1989	3	1.7×10^{-3}	3.0×10^{12}	1.7	5.4×10^{-4}
1990	5	2.8×10^{-3}	5.0×10^{12}	2.8	9.0×10^{-4}
1991	3	1.7×10^{-3}	3.0×10^{12}	1.7	5.4×10^{-4}
1992	1	5.5×10^{-4}	1.0×10^{12}	0.7	1.8×10^{-4}
1993	<1	$<5.5 \times 10^{-4}$	$<1.0 \times 10^{12}$	<0.8	$<1.8 \times 10^{-4}$
1994	3	1.7×10^{-3}	3.0×10^{12}	4.0	5.4×10^{-4}
1995	1	5.5×10^{-4}	1.0×10^{12}	0.8	1.8×10^{-4}
1996	7	3.8×10^{-3}	7.0×10^{12}	4.8	1.3×10^{-3}
1997	9	5.0×10^{-3}	9.0×10^{12}	7.2	1.6×10^{-3}
1998	5	2.9×10^{-3}	5.0×10^{12}	3.6	9.0×10^{-4}
1999	11	6.1×10^{-3}	1.1×10^{13}	11.0	2.0×10^{-3}
2000	7	3.9×10^{-3}	7.0×10^{12}	8.2	1.3×10^{-3}
2001	9	5.0×10^{-3}	9.0×10^{12}	7.9	1.6×10^{-3}
2002	9	5.0×10^{-3}	9.0×10^{12}	6.1	1.6×10^{-3}

The electricity production data are from PRIS.
[a] Unit 1 put into operation in December 1983.
[b] Unit 2 put into operation in August 1987.

The ^{14}C value in the very thin soil sample (less than 1 cm) below the moss from the pier of cooling water inlet channel was found to be significantly higher than in any of the vegetation samples, with the ^{14}C specific activity up to 813 pMC (sample ID ICB-s: cooling water inlet channel, sampling point B, soil with humus). This pier itself is made of large concrete blocks that have never been flooded by lake water. The horizontally (or close to this orientation) located concrete blocks in course of time (several decades) are naturally covered by thin layer of deposited mineral and organic particles (soil with humus) and moss. The ^{14}C excess in the samples from the pier of heated water outlet channel (samples ID OCA, OCB) was significantly lower – up to 205 pMC for moss, and 96 pMC for soil. Furthermore, the pier of heated water outlet channel was several times reconstructed and does not contain concrete surfaces with such long exposition as pier of cooling water inlet channel. A ^{14}C specific

Table 3
The ^{14}C specific activity in the moss and soil samples collected within 500-m distance from the Ignalina NPP on 21 June 2005

Sample ID	Sample type and location	^{14}C specific activity (pMC $\pm 1\sigma$)	^{14}C excess (pMC $\pm 1\sigma$)
Background	Moss, the Vilnius district, southeastern Lithuania	107 ± 2	
	Moss, near the INPP (1 km to the SW)	141 ± 2	34 ± 3
ICA	Moss, cooling water inlet channel, point A	115 ± 4	8 ± 4
ICB-m	Moss, cooling water inlet channel, point B	564 ± 7	457 ± 7
ICB-s	Soil with moss remains, inlet channel, point B	813 ± 10	706 ± 10
OCA-m	Moss, heated water outlet channel, point A	312 ± 4	205 ± 4
OCB-m	Moss, heated water outlet channel, point B	167 ± 2	60 ± 3
OCB-s	Soil with moss remains, heated water outlet channel, point B	203 ± 2	96 ± 3

Sampling sites are shown in Fig. 2. The contemporary background of 106.7 pMC for 2005 was measured in moss from the Vilnius district, southeastern Lithuania. The value 106.7 pMC corresponds to volumetric activity of ^{14}C in air equal to 0.043 Bq m^{-3} assuming that the CO_2 concentration in air is 330 ppm. In each sampling site one (moss) or two samples (moss and related soil) were taken. Only statistical radiometric uncertainty (1σ) is reported.

activity of 2310 pMC in soil samples from the pier of cooling water inlet channel was reported in some places in Magnusson et al. (2004, 2007). In latter study, the ^{14}C excess was believed to be caused by releases of particulate material. Due to uneven and rare spots with a high specific activity of ^{14}C in soil, it seems unlikely that the high ^{14}C excess in the pier samples would be caused by high release rates of $^{14}CO_2$ or even of ^{14}C with particulates from the main stacks in the past. The release from stacks of 150 m height would form uniform and wide ^{14}C trace on the earth surface according to Gaussian plume model. One of the potential sources of particulate material could be equipment for processing of radioactive waste with stacks of 75 and 12–13 m heights, however, this assumption based on available information is not justified yet.

Because of the distance from the pier samples to the water level (more than 1.5 m) and the moderate ^{14}C content in aquatic plants, which has been reported to range mainly from 121 to 155 pMC (Mikhajlov et al., 1999; Jakimaviciute-Maseliene et al., 2003; Magnusson et al., 2007), waterborne ^{14}C releases have not been considered as a significant source of pier contamination. As the ^{14}C concentrations found in the moss and related soil samples in very limited area of the Ignalina NPP environment are not diluted and extremely high, the possibility of airborne ^{14}C particulates must be considered as discussed by Marsden et al. (2002) and Magnusson et al. (2007).

5. Conclusions

The ^{14}C concentration near the Ignalina NPP has been studied by analysing annual tree rings, moss and related soil samples using conventional liquid scintillation counting. A comparison between the specific activity of ^{14}C in the tree rings near the Ignalina NPP and specific activity of the tree rings from the background regions has revealed the ^{14}C excess over the whole operation period of the Ignalina NPP (1984–2002). Based on the experimentally-determined ^{14}C excess value, the maximal value of normalized release rate (11 TBq GW_e^{-1} $year^{-1}$) was estimated over this time span. In comparison with the other radionuclide releases from the Ignalina NPP and respective doses (Motiejunas et al., 1999), the effective dose resulting from the ^{14}C is the highest reaching 2.0×10^{-3} mSv $year^{-1}$.

The excess of ^{14}C specific activity measured in the moss and related humus samples from moss-covered sites (concrete blocks) within short distances (up to 0.5 km) of the power plant show highly elevated ^{14}C contents, probably indicating releases of particulate material, as previously discussed in Magnusson et al. (2007). The ^{14}C excess farther from nuclear power plant (0.5–30 km zone and beyond 30 km zone) is insignificant.

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