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Carbon-14 in Terrestrial and Aquatic Environment of Ignalina Nuclear Power Plant: Sources of Production, Releases and Dose Estimates

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1. Introduction

The development history of nuclear power in world already has over passed the limit of 50 years. This time span was sufficiently long for many nuclear reactors to complete their operation stage and to enter the decommissioning stage. The Ignalina NPP (INPP), Lithuania, is one of them. Its operation history only lasted for 26 years for different reasons but mainly the political ones. The INPP consists of two RBMK-1500 reactor units, Unit 1 and Unit 2 (Almenas et al., 1998). 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 with the principle of electricity generation the same as for boiling water reactors (BWRs).

The Ignalina NPP is located in the north-eastern part of Lithuania near the borders of Belarus and Latvia (Fig. 1). The NPP uses the Lake Druksiai as a natural reservoir for cooling water. 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. The Lake Druksiai is a flow-through lake with six small creeks flowing in and one river, with a water regulation dam, flowing out.

The two reactor units, Unit 1 and Unit 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 was 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 on December 31, 2009. During the 26 years of operation, the INPP has produced 307.9 billion kWh of electricity: Unit 1 – 136.9 billion kWh and Unit 2 – 170.2 billion kWh. The total amount of electricity sold is 279.8 billion kWh (http://www.iae.lt). The INPP operational history and the routine radiation in environment monitoring data evidenced that INPP was operated safely and helpfully for the society.

The routine monitoring of radiation in environment of NPPs often does not include some important nuclides, namely carbon-14 (^{14}C), which have or may have significant contribution to effective dose of human exposure in the whole nuclear fuel cycle. [14]C (half-life 5730 years, maximum beta particles energy of 156 keV) occurs naturally in the environment due to the cosmic-ray (secondary neutrons) induced production in the lower stratosphere and upper troposphere on nitrogen atoms ([14]N(n,p)[14]C) (Libby, 1946; Raaen et al., 1968). The naturally occurring cosmogenic ^{14}C is generated at a rate of 1.5×10^{15} Bq/year.

Fig. 1. Study area location

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 and consequent anthropogenic emissions via the Suess effect (Fairhall & Young, 1970). In the 1950s, when many atmospheric nuclear weapon tests took place, the ^{14}C concentration in the air rose sharply to a maximum (double the natural concentration), and gradually decreased in 1960s and later after cessation of atmospheric tests. The ^{14}C activity in the atmosphere has decreased steadily due to CO_2 uptake by the oceans and by the biosphere (Hertelendi & Csongor, 1982; Levin & Kromer, 2004). [14]C is also artificially produced in all types of nuclear reactors by the similar neutron-induced reactions on 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). 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).

[14]C is referred to as one of the difficult-to-measure nuclides (pure beta emitters) due to the presence of other radionuclides in a sample. [14]C is rarely measured in process media of nuclear reactors and even in gaseous releases from nuclear plants notwithstanding that equipment for monitoring ^{14}C in gaseous releases is today commercially available. However,

releases of gaseous ^{14}C from nuclear power reactors result in prevailing dose fraction compared to all radionuclides. This is a case for the INPP (Nedveckaite et al., 2000).

In 2002, the Government of Lithuania approved the adoption of the ''immediate dismantling'' strategy for decommissioning of both INPP units. A key component of this decommissioning strategy is to dispose of operating and decommissioning waste in a near-surface repository (NSR). ^{14}C as a long-lived and mobile in the environment radionuclide and due to large inventory has been recognized as one of the most important nuclides in the assessments of doses for future generations arising from INPP NSR for low- and intermediate-level, short-lived radioactive waste (LILW) disposal. Based on the scaling factor method (Lukauskas et al., 2006; Plukis et al., 2008), a preliminary inventory of ^{14}C for NSR with capacity for disposal of approximately 100 000 m^3 of waste was evaluated as 1.43×10^{13} Bq. Similar figures of ^{14}C inventory were evaluated for NSRs in other countries: Japan, Rokkasho-Mura disposal facility, first 40 000 m^3, maximal inventory - 3.37×10^{12}, Spain, El Cabril disposal facility, maximum permissible activity - 2.00×10^{13} Bq (IAEA, 2004).

From radioactive waste long-term management point of view, the importance of ^{14}C is also due to the large total mass of graphite per RBMK-1500 reactor (more than 1800 tons) and consequently there is a large ^{14}C inventory in graphite (IAEA, 2006). Based on calculations of graphite impurities activation, ^{14}C and tritium contribute mostly (respectively 3.91×10^{14} Bq and 2.19×10^{14} Bq) to the total activity of all the irradiated graphite constructions of the reactor (Ancius et al., 2004). It is the rough upper limit estimate, derived assuming that activation products are retained in the graphite. Activated graphite of INPP due to the high specific activity (up to 5×10^5 Bq/g) of the long-lived ^{14}C is classified as the long-lived radioactive waste.

The worldwide nuclear power operational experience gives evidence that ^{14}C is continuously released to environment from NPPs, is key radionuclide of NSRs for LILW disposal, forms significant fraction of irradiated graphite radionuclide inventory, retains in the spent nuclear fuel and consequently will be disposed of in geological repositories for long-lived high activity waste in the form of spent fuel or radioactive waste arising from spent fuel reprocessing. In this sense, we can consider ^{14}C as one of the most powerful environmental tracers of nuclear fuel cycle. Because of the biological importance of carbon (^{12}C, ^{13}C and ^{14}C) and, consequently, the biological incorporation of radioactive ^{14}C through photosynthesis, it is of great interest to run ^{14}C measurements in environment, especially if ^{14}C is not monitored in releases as was a case in the INPP operational history. Considering that ^{14}C specific activities of terrestrial and aquatic plants may vary according to the atmospheric ^{14}C/^{12}C ratio, many studies have investigated atmospheric and sometimes aquatic ^{14}C activities in the vicinities of nuclear power plants using vegetation as bioindicators (Obelic et al., 1986; Levin et al., 1988; Loosli & Oeschger, 1989; Buzinny et al., 1995; Milton et al., 1995; Milton & Kramer, 1998; Stenström et al., 1996; Mikhajlov et al., 1999; Magnusson et al., 2004; Mazeika et al., 2008).

The main aim of this study is to investigate how terrestrial and aquatic vegetation (organic carbon, Corg) and dissolved inorganic carbon (DIC) of aquatic environment recorded the ^{14}C activity excess compared to the contemporary ^{14}C background level during the whole period of the INPP operation (the end of 1983–the end of 2009). The ^{14}C activity excess data allow evaluating the order of magnitude of gaseous and liquid release rates due to operation of the NPP what can partially substitute direct ^{14}C release monitoring data and give basis for approximate dose estimates. The other goals are: to give closer look on ^{14}C activity aerial

dynamics in terrestrial environments (in 1996 and 2001); to present first experimental data on ^{14}C activities in different process water systems of the INPP reactor and in various water clean-up systems, namely, in evaporator concentrates and spent ion-exchange resins (taken in 2002). This chapter mostly presents the summarization of $^{14}CO_2$ specific activities in terrestrial and aquatic plants and $H^{14}CO_3^-$ specific activities in aquatic environment measured during various research projects performed in the INPP vicinity.

2. Methods

2.1 ^{14}C production in nuclear reactors

The normal operation of NPPs produces various radionuclides by fission within the fuel or by neutron activation in the structural materials and component systems of the reactor. The escape of these radionuclides from the reactor and its auxiliary process systems during operation time generates a variety of solid, liquid and gaseous radioactive waste. Despite that the design of the reactor ensures minimized escaping of radionuclides from technological systems, some radionuclides, namely ^{14}C, are continuously discharged in various effluents to environment.

The major ^{14}C producing neutron activation reactions in NPP reactors are (IAEA, 2004; Buchuev et al., 2002):

(a) The $^{14}N(n,p)^{14}C$ reaction with a very high thermal neutron capture cross-section (1.82 barn (1 barn = 10^{-24} cm^2)); (b) The $^{17}O(n, \alpha)^{14}C$ reaction with a high thermal neutron capture cross-section (0.24 barn); (c) The $^{13}C(n, \gamma)^{14}C$ reaction with a low cross-section (0.9×10^{-3} barn); (d) The $^{15}N(n,d)^{14}C$ reaction with a very low cross-section (2.5×10^{-7} barn); (e) The $^{16}O(n,^3He)^{14}C$ reaction with a very low cross-section (5.0×10^{-8} barn).

Most of ^{14}C is produced in nuclear power reactors by $^{14}N(n,p)^{14}C$ reactions with nitrogen in fuels, moderators and coolants as a primary impurity, by $^{17}O(n, \alpha)^{14}C$ reactions in oxide fuels, moderators and coolants, and by $^{13}C(n,\gamma)^{14}C$ reactions in graphite moderators. Reactions (a) (b) and (c) are the most important contributors to ^{14}C production in thermal reactors. ^{14}C is also a ternary fission product, but the amount produced in this way is negligible.

The substrate atoms for the activation reactions (i.e. nitrogen, oxygen and carbon) occur widely in fuel, and in cladding, moderator, coolant or structural material, either as major constituents or as impurities. In consequence, ^{14}C produced in a reactor can be released directly to the environment from the coolant and/or moderator in a gaseous form or in much smaller quantities as liquid effluents. ^{14}C can remain in the reactor core until a reactor is decommissioned (e.g. in the graphite moderator of RBMK reactors). In INPP the spent fuel is stored in dry interim storage facility as long as 50 years before the final disposal.

^{14}C is produced in the fuel and coolant and is distributed wherever gas or fluid streams take place in NPP. Leakage of NPP systems allows an eventual release to the environment. As it is reported in (NCRP, 1985; Milton et al., 1995; IAEA, 2004), heavy water reactors (CANDU) and RBMK reactors have highest ^{14}C release rates compared to other types of reactors. The estimated ^{14}C release rate for 14 operating RBMK reactors in East Europe is 1.85×10^{12} Bq/year per reactor. The normalized emission level of ^{14}C from the RBMK type reactor in the Russian Federation has been estimated to be 1.0 ± 0.3 TBq×GW(e)$^{-1}$×year^{-1} (Gaiko et al., 1985).

The following systems have been considered to be release pathways for gaseous ^{14}C from a BWR (IAEA, 2004):

(a) The condenser steam jet air ejector; (b) The turbine gland seal condenser exhaust; (c) The reactor building purge exhaust; (d) The turbine building ventilation system exhaust; (e) The radioactive waste building ventilation system exhaust.

The condenser steam jet air ejector is expected to be the most significant release point (>99% of total ^{14}C release).

A simplified scheme of main ^{14}C flows based on radioactive waste generation streams in INPP (Plukis et al., 2008) is presented in Fig. 2.
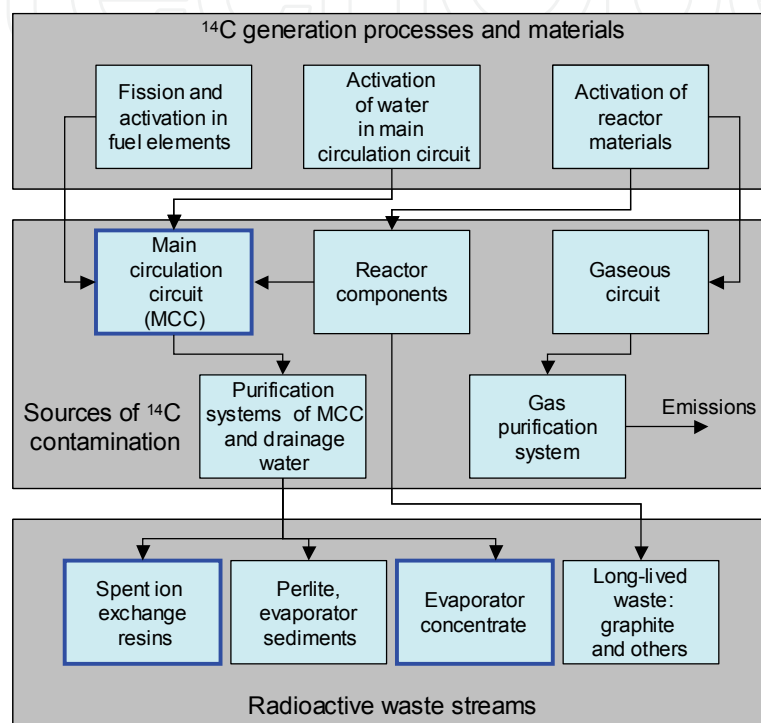

Fig. 2. A simplified scheme of main ^{14}C flows in INPP. Blue borders show studied reactor water and radioactive waste objects

^{14}C can enter the main circulation circuit (MCC) coolant (reactor water) of INPP due to fuel cladding defects, corrosion of metal structures of reactor core components and direct generation in the coolant. The ^{14}C from contaminated coolant of MCC can be further transferred to the final waste or to environment by three main routes: (i) the retention of ^{14}C in structural materials of NPP, (ii) the loss of a coolant through leakages to the drainage system, and (iii) the chemical purification of a coolant by ion exchange resins and perlite. The design of RBMK with only one circuit leads to a continuous release of ^{14}C through the ventilation stack.

2.2 Sampling and measurements

^{14}C in the INPP environment has been studied in terrestrial and aquatic plants, annual tree radial growth rings, and surface water and, at a smaller scale, in unconfined groundwater in various research projects during the whole operation period of INPP.

The main biota samples, including tree rings, as well as water samples from surface water bodies and groundwater monitoring wells (up to 20 m depth) were collected at a short distance (0.5-2 km) from the power plant (Figs. 3-4).
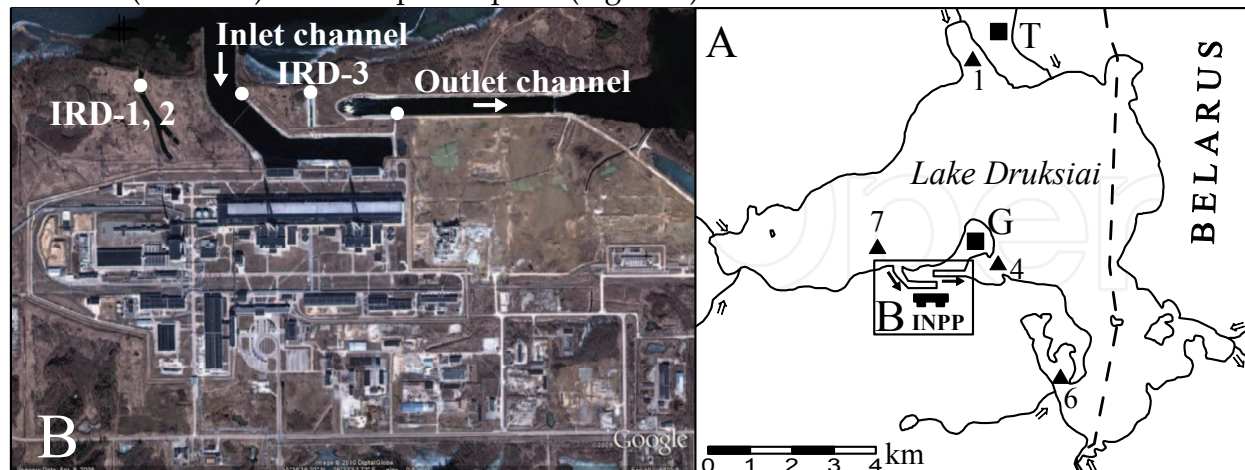

Fig. 3. Surface water and biota sampling points in Lake Druksiai and in terrestrial environment (A), and in the main channels related to INPP industrial site (B)
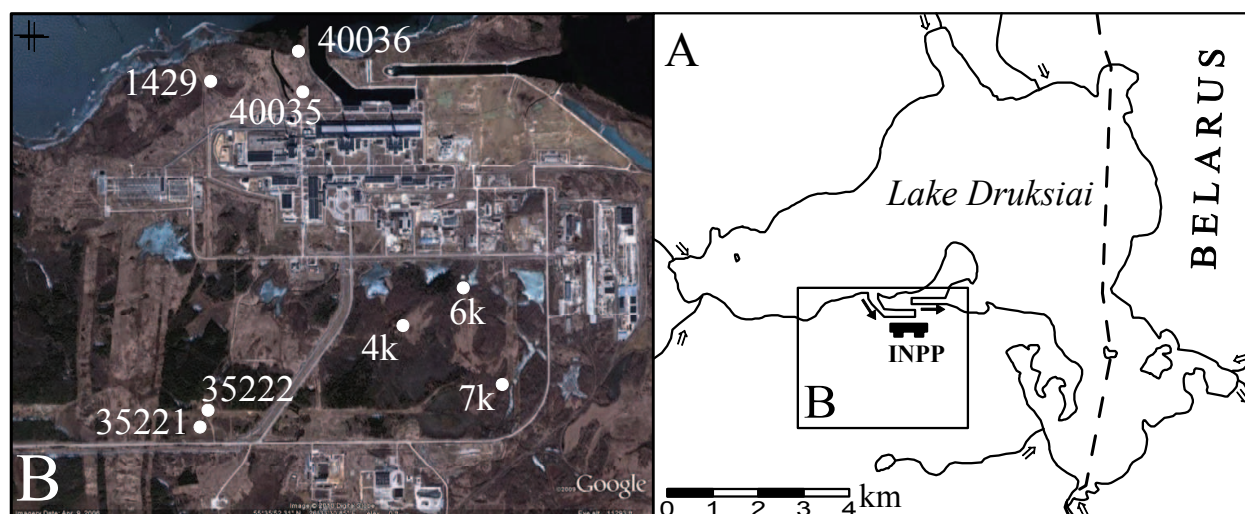

Fig. 4. Groundwater sampling points close to INPP industrial site (B)

Most often terrestrial plant samples were collected from two areas: Tilze (T monitoring site) and Grikiniskes (G monitoring site). T site is attributed to background area where impact of INPP is low and negligible. G site is located in the so called "impact zone" which basing on simple semi-empirical Gausian plume model for estimating long-term average radionuclide concentrations in air (Gusev & Beliajev, 1986; Metodology..., 1995) was considered as mostly affected by airborne release from the INPP. Terrestrial plant samples from wider area were collected in 1996 (wormwood stems) and in 2001 (alder leaves) at a longer distance (up to 30 km) from the INPP. The terrestrial plant samples for recent years included the following species: European blueberry, Kuhn brake, Roth. rough smallreed, Hull common heather, Shimp stair-step moss and others. The aquatic samples were of macrophytes *Myriophyllum spicatum*, *Ceratophyllum demersum* and filamentous algae mainly.

Carbon-14 in Terrestrial and Aquatic Environment of Ignalina Nuclear
Power Plant: Sources of Production, Releases and Dose Estimates
299

Several cross-sections of the trunk of pine trees from the background regions of Lithuania and "impact zone" for separating the annual tree rings were taken in 1996, 2002 and 2003.

The water sampling by precipitating DIC from the water for ^{14}C activity concentration in the Lake Druksiai measurement have been already started before the INPP operation (Mazeika, 2002). After the start of INPP operation the new monitoring points on surface water bodies related to the INPP industrial site were established: cooling water inlet channel (IC), heated water outlet channel (OC), industrial discharge and rain water drainage (IRD) channels 1, 2, and 3 (Fig. 3).

The measurements of ^{14}C activity concentration in DIC of groundwater started during the INPP pre-operation period. In the course of time, the general view of groundwater monitoring network has changed. The network of recent years is shown in Fig. 4 (the groundwater monitoring network on site which is operated by the INPP is not included).

Groundwater sampling is mainly attributed to three monitoring well groups. The first group of wells with numbers 1429, 40035 and 40036 is installed in the "impact zone" on groundwater flow pathway down the INNP. Well No 1429 is very shallow with screening interval of 3–5 m. It has the longest period of sampling and is directly related to operational radioactive waste storage area. Wells No 40035 and 40036 with screening interval of 18-20 m are located close to groundwater flow draining boundaries (Lake Druksiai and channels IRD-1 and 2). These wells capture more diluted groundwater flow path lines compared to well 1429. The second group of wells with numbers 35221 and 35222 is not related directly to the INPP site. However these wells with screening interval of 10-20 m are installed in the quarry where the sludge from the Visaginas wastewater treatment plant was disposed of in the period preceding the quarry re-cultivation. An interest (from the point of view of radiation protection) to this area has occurred due to the fact that domestic wastewater from the INPP (from shower and laundry facilities, etc.) was treated at the same wastewater treatment plant. The third group of wells with numbers 4k, 6k and 7k are attributed to the "background area" as they are on the groundwater flow pathway upwards the INPP.

Vegetation and tree annual ring samples (up to 50 g) were dried, pyrolised in no oxygen atmosphere. The charred material was left for further procedures. Dried carbonates (up to 150 g) precipitated in field from surface and ground water samples in alkaline media adding NaOH and $CaCl_2$ into water were converted to CO_2 by treatment with HCl. The collected CO_2 was stored for further procedures.

The specific activity of ^{14}C in environmental samples was measured by liquid scintillation counting (LSC) method (Gupta & Polach, 1985; Arslanov, 1985). A conventional procedure for benzene synthesis was applied (Kovaliukh & Skripkin, 1994). The several steps were followed: (1) reaction of carbon containing sample material (CO_2 for DIC of water and charred carbon for vegetation samples) with Li in the metallic reactor under vacuum at the temperature 600-700°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 H_2SO_4 and by vacuum sublimation, and (5) adding the scintillation admixtures, PPO and POPOP, to benzene produced from sample material. 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±0.8%; and Figure of Merit (Efficiency^2/Background) of 12 380±700.

The ^{14}C results were presented as specific activity in Bq/kg C or in percent of modern carbon, pMC (1 pMC – 2.26 Bq/kg C) (Stuiver & Polach, 1977). As bicarbonate ion is the main chemical form of ^{14}C in the water, the ^{14}C activity concentration in Bq/m^3 is calculated from two experimentally measured values such as HCO$_3^-$ concentration and ^{14}C specific activity relative to carbon. As concentration of bicarbonate ion in the surface water is relatively constant, the both units pMC and Bq/m^3, express the same variation figure in time. However in the course of time, the concentration of bicarbonate ion in the groundwater ranges significantly in closely located wells or even in the same well. For groundwater it is useful to compare ^{14}C data expressed in both units, in pMC as is common for isotope geochemistry and dating issue, and in Bq/m^3 what is essential for radiation protection issue.

One limited sampling campaign in 2002 was aimed to measure ^{14}C activity in different process water systems of the reactor and in various water clean-up systems, namely, in evaporator concentrates and spent ion-exchange resins. The obtained data together with other available data (Plukis et al., 2008) were used for statistical analysis of radionuclide specific activities in the INPP technological media and radioactive waste and for the calculation of the corresponding scaling factors. Correlations of difficult-to-measure radionuclides to the key radionuclide ^{60}Co showed that the scaling factor method could be applied to the RBMK–1500 reactor waste characterization.

The samples from the process water systems of the reactor and various water purification systems were provided by the INPP staff. The main processes of ^{14}C extraction from liquid samples were acid stripping and wet oxidation (Veres et al., 1995; Uchrin et al., 1998) transferring dissolved inorganic carbon (DIC) and dissolved organic carbon (DOC) into ^{14}CO$_2$, while the latter one was precipitated in the form of BaCO$_3$. For ion-exchange resins, an initial amount of material (2 g) was boiled in the 5-8% NaOH over 1-2 hours in order to transfer the groups of anions into solution which was separated from resins and treated with the sulphuric acid and hydrogen peroxide in the same way as for other liquid samples. Chemical yield of ^{14}C extraction was not less than 80%. Barium carbonate was mixed with selective carbon dioxide absorbent and scintillation cocktail. ^{14}C activity in the resulting solution was measured by the LSC method. ^{14}C counting efficiency was in range of 20-40% and was estimated on BaCO$_3$ sample with the known ^{14}C activity. This sample was prepared from the standard sodium carbonate (Na$_2$CO$_3$) solution.

The uncertainty of the ^{14}C estimations (statistical radiometric error) was reported at 2 and 1 sigma-level for the tree rings samples and for all other studied samples, respectively. The quality of ^{14}C determinations in our laboratory was sufficiently reliable what was periodically tested through participating in various inter-comparisons. The tests indicated that most of the low activity ^{14}C determinations are of high (1.5%) precision. Lower precision (up to 15%) of the ^{14}C determinations is evident for higher activity samples from the INPP process media.

3. Results and discussion

The main data on ^{14}C in terrestrial and aquatic plants, in DIC of surface water and groundwater for the whole period of observations are summarized in Figs 2-9. Some new ^{14}C data attributed to wider species diversity of terrestrial and aquatic plants for recent period of observations (2007-2009) are in detail presented in Tables 1 and 2.

3.1 ^{14}C in the terrestrial environment

The ^{14}C specific activity in the terrestrial annual vegetation from the "impact zone" of INPP was compared with the ^{14}C activity in the local background areas and in the air of the Northern Hemisphere for the period 1983-2008 (Fig. 5).
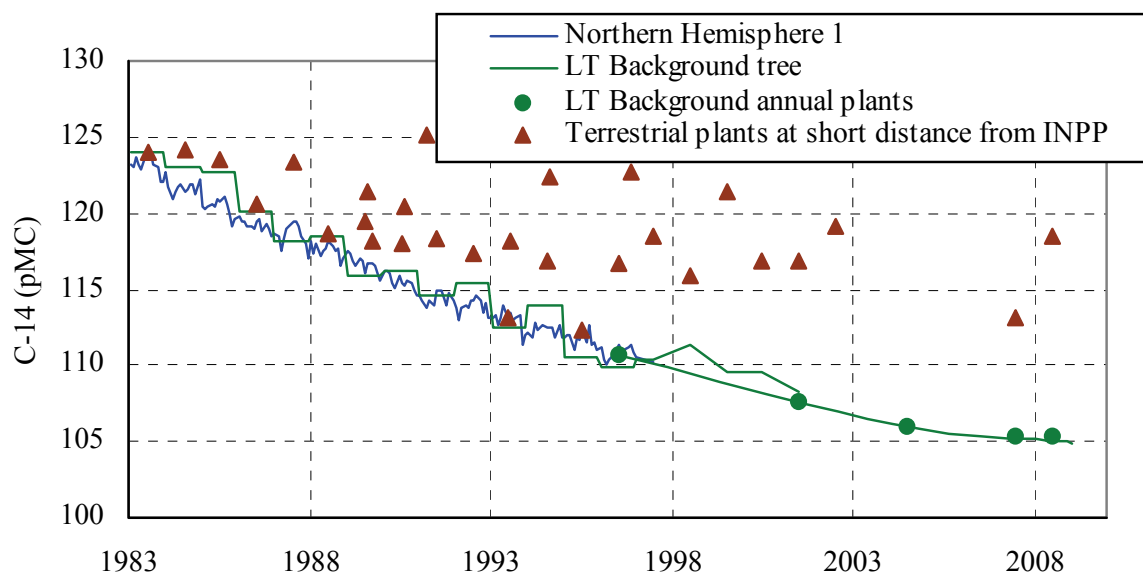

Fig. 5. The ^{14}C specific activity of terrestrial plants at short distance from INPP for the period of INPP operation 1983–2008, compared with ^{14}C activity in air of Northern Hemisphere according to Hua & Barbetti (2004) and with local background ^{14}C level

^{14}C background plots were built from several data sets: Northern Hemisphere data for the period 1983–1997 (Hua & Barbetti, 2004), local tree rings data (Mazeika et al., 2008) and data on annual vegetation from the Lithuanian national parks. All three data sets are in good correlation and show the continuous ^{14}C of thermonuclear weapon tests to decline from 124 pMC in 1983 to 106 pMC in 2008. Nowadays, the ^{14}C background is approaching the ^{14}C level of cosmogenic origin. Most of ^{14}C data for the "impact zone" of INPP are clearly above the background level with maximal excess of 10-15 pMC for 1988–2008. Evident variability of ^{14}C excess is related to aerial position of samples.

The aerial distribution of ^{14}C activity in the annual vegetation was analyzed twice – in 1996 and in 2001. Specific activity of ^{14}C in wormwood stems (12 samples) ranged from 96 to 123 pMC in 1996, and from 107 to 178 pMC in alder leaves (23 samples) in 2001. The lowest ^{14}C values were in line with the background ones, and the highest values gave the same ^{14}C excess of 10-15 pmC. However one sample of 2001 had ^{14}C excess up to 70 pMC. In both cases, the highest specific activity of ^{14}C was detected in the zone north-east of the INPP. This "impact zone" coincides with the dominant wind direction.

The analyzed data are in line with ^{14}C in tree rings analysis (Mazeika et al., 2008), which revealed a ^{14}C excess in tree rings near the INPP in the same area. At the beginning of the INPP operation, the ^{14}C excess was negligible whereas in 1996–2002, the ^{14}C excess value was up to 5-11 pMC. For the period of 2003–2006 the data are lacking however judging by

[14]C data for 2007–2008 there is an evidence of ^{14}C excess in the same area - up to 10 pMC level (Table 1).

Sampling point location	Species	Sampling date	[14]C (pMC±1σ)
INPP region, Grikiniskes (G locality in Fig. 3), "impact zone " of INNP	European blueberry	28-06-2007	110.1±0.9
	Kuhn brake	28-06-2007	108.2±1.3
	Roth. rough smallreed	28-06-2007	108.7±1.1
	Roth. rough smallreed	28-08-2008	110.8±0.5
	Hull common heather	28-06-2007	113.2±1.3
	Hull common heather	28-08-2008	112.7±0.5
	Shimp stair-step moss	28-08-2008	118.5±0.5
Zarasai district, Tilze (T locality in Fig. 3), local background	Shimp stair-step moss	28-06-2007	104.7±1.1
	Kuhn brake	28-06-2007	104.5±1.3
	Shimp stair-step moss	29-08-2008	104.6±0.5
	Hull common heather	29-08-2008	104.9±0.5
	Roth. rough smallreed	29-08-2008	106.5±0.5
Lithuanian national parks (not shown)	European blueberry	28-06-2007	101.4±0.8
	Kuhn brake	01-08-2007	102.7±1.3
	Hull common heather	01-08-2007	105.4±1.2

Table 1. [14]C specific activity in terrestrial plants from INPP "impact zone" and background areas in 2007-2008

Following the approach presented in (Mazeika et al., 2008) we can again support the estimation that maximal ^{14}C release rate from the INPP is of order 10^{13} Bq/year. In the case of an atmospheric release, the nuclide-specific routine release conversion factors, representing the ratio of the annual effective dose for a critical group member (Sv/year) at the location of the highest predicted radionuclide concentration in air, to the activity released from the INPP (Bq/year), are set (Nedveckaite et al., 2000; LAND, 42–2001). The effective dose resulting from the ^{14}C mentioned release rate is 2.0×10^{-3} mSv/year.

3.2 ^{14}C in the aquatic environment

In the aquatic environment, ^{14}C basically may be involved into two main components: into the water as part of dissolved bicarbonate ions, HCO_3^-, and into the plant tissue as Corg. Both chemical forms of ^{14}C are directly or indirectly related to atmospheric CO_2. Therefore in the lakes with short water residence in the system time ^{14}C activity in both chemical forms is similar. As the significant area of Lake Druksiai belongs to airborne release from the INPP impact zone, the atmospheric [14]CO_2 may slightly enhance the ^{14}C level. If ^{14}C is released to the lake with liquid effluents in inorganic and organic chemical forms it can be traced investigating both DIC of water and Corg of macrophytes as both carbon compartments are interacting in biogeochemical cycle of aquatic ecosystem.

[14]C activity measurements in DIC of several surface water bodies with different frequency have been performed since 1975 (Fig. 6).
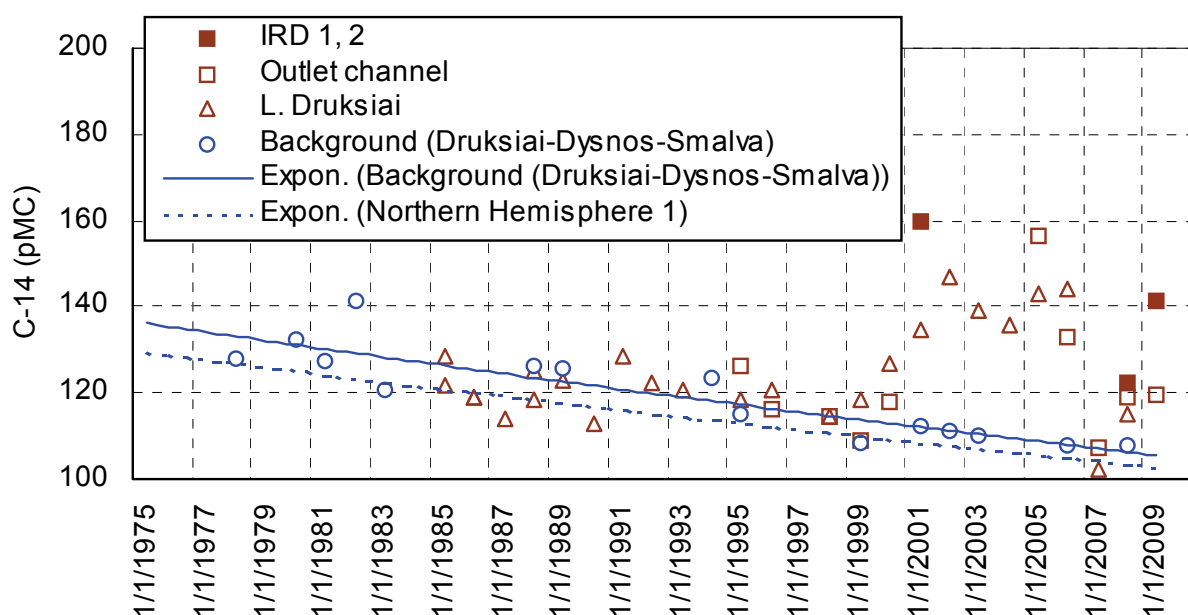
Fig. 6. The ^{14}C specific activity of DIC in water bodies related to INPP at short distance from INPP for the period 1975–2009, compared with ^{14}C activity in air of Northern Hemisphere according to Hua & Barbetti (2004) and with local background ^{14}C level

Several data sets were used as ^{14}C background: the Northern Hemisphere data for the period 1975–1997, the data on the Lake Druksiai before the INPP operation, and the data on the Lake Dysnos and rivulet Smalva during the INPP operation. [14]C activity trend lines generated from the Northern Hemisphere data and local lakes data are very comparable.

For a significant period (1983–1999) of ^{14}C monitoring in DIC of surface water, the influence of INPP has been hardly recognized. [14]C activity in DIC from the Lake Druksiai and cooling water inlet channel has increased in 2002–2006 with the ^{14}C excess of 30–35 pMC above the background level. The similar ^{14}C activity trend was observed in the technical water inlet channel, which represented ^{14}C activity diluted and averaged to almost all lake water volume. The IRD 1, 2 channel is a water body with the lowest diluted effluents and enhanced ^{14}C activity in DIC in 2001 (159 pMC) and 2009 (142 pMC). The traceable ^{14}C excess in DIC of the water from the Lake Druksiai allows evaluating the order of ^{14}C release rate from the INPP to the water of $\sim 10^8$ Bq/year using radionuclides activity dilution factors derived in (Mazeika & Motiejunas, 2003). The effective dose resulting from the ^{14}C mentioned release rate is $\sim 3.0 \times 10^{-4}$ mSv/year. [14]C in aquatic plants from the Lake Druksiai was measured occasionally and most systematically in resent years (Fig. 7, Table 2).

For ^{14}C excess evaluation, the same background data sets as for terrestrial plants were used. The evident ^{14}C excess in aquatic plants with significant variability for the period 1995-2008 is the main finding. The highest ^{14}C values (up to 172 pMC) were observed in the lake area close to monitoring station No 7 (Fig. 3) where the effluents discharged by IRD 1, 2 channel (maximum ^{14}C activity in biota in 2007 was 263 pMC) were interacting with plants in short distance before diluting and averaging in lake water masses. The macrophytes were recognized as good bioindicators to show a short-term ^{14}C variation during the vegetation

season. [14]C data on DIC of water and Corg of plants in the aquatic environment are in agreement and complements each other.

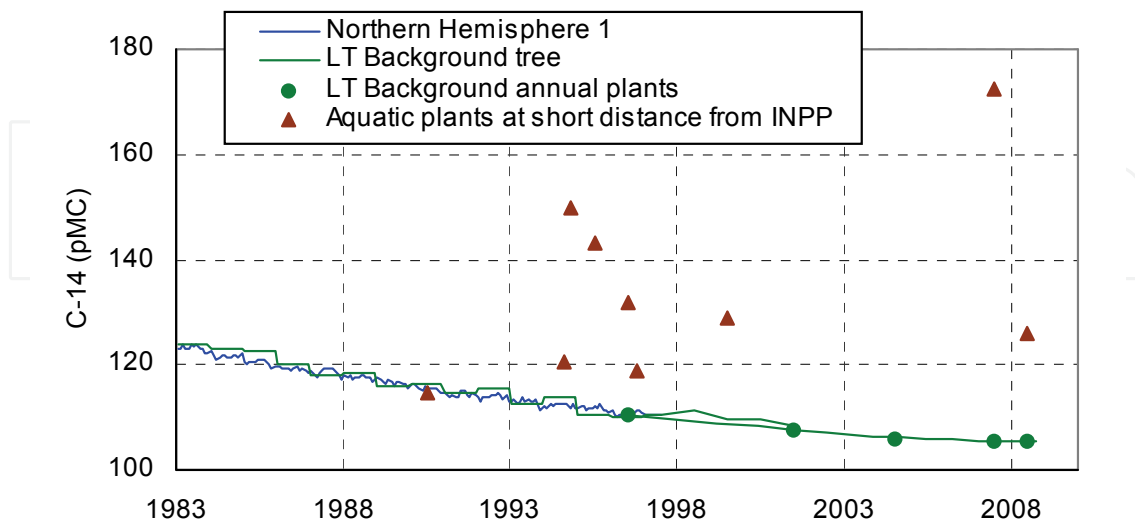Fig. 7. The ^{14}C specific activity of aquatic plants at short distance from INPP, compared with ^{14}C activity in air of Northern Hemisphere according to Hua & Barbetti (2004) and with local background ^{14}C level

Sampling point location (See location on Fig. 3)		Species	Sampling date	[14]C (pMC±1σ)
Technical water from INPP outlet channel (OC)		*Myriophyllum spicatum*	27-06-2007	108.8±1.6
			28-08-2008	127.7±0.6
Drainage channel (IRD-1, 2)		*Ceratophyllum demersum*	27-06-2007	262.6±2.0
			28-08-2008	172.9±7.7
Lake Druksiai	1 station	Filamentous algae	27-06-2007	111.1±0.9
		Myriophyllum spicatum	27-06-2007	119.3±1.9
		Ceratophyllum demersum	24-09-2008	118.8±1.0
	4 station	*Ceratophyllum demersum*	27-06-2007	111.6±1.5
		Myriophyllum spicatum	27-08-2008	125.9±0.7
	6 station	*Ceratophyllum demersum*	27-06-2007	106.3±1.5
	7 station	*Ceratophyllum demersum*	27-06-2007	172.3±2.8
			27-08-2008	123.4±0.9

Table 2. [14]C specific activity in aquatic plants from Lake Druksiai and channels related to INPP industrial site in 2007-2008

Carbon-14 in Terrestrial and Aquatic Environment of Ignalina Nuclear
Power Plant: Sources of Production, Releases and Dose Estimates
305

3.3 ^{14}C in the groundwater

The main chemical form of ^{14}C in the groundwater is bicarbonate ions or more generally defining DIC. A small fraction may be in organic carbon. Both chemical forms of ^{14}C in the groundwater originate in the soil–unsaturated zone system through the carbon biogeochemical cycle. They are related to atmospheric CO_2 and have often the same or similar ^{14}C specific activity respect to carbon. Even if these specific activities are equal, the volumetric activity of ^{14}C in the water from DIC will be higher then that from organic carbon due to significant differences in chemical concentrations of those two components in common groundwater. ^{14}C specific activity in the groundwater with short residence time usually is close to that of the atmospheric CO_2. There are often cases when aquifer matrix involves carbonate material, groundwater interacts with matrix long time and by presence of CO_2 surplus transfers HCO_3^- to the water from solid "old" carbonates not containing ^{14}C. These processes of ^{14}C geochemical dilution in the groundwater are well studied in the isotope hydrology (Clark & Fritz, 1997). In such cases, ^{14}C specific activity in DIC of groundwater may be significantly lower then in the atmospheric CO_2.

If ^{14}C is released to the groundwater with liquid effluents in inorganic and organic chemical forms it can be traced investigating firstly DIC of groundwater. For 1987–2009, ^{14}C specific activity of DIC in the water (pMC) and ^{14}C volumetric activity concentration (in Bq/m^3) are presented in Figs. 8, 9.
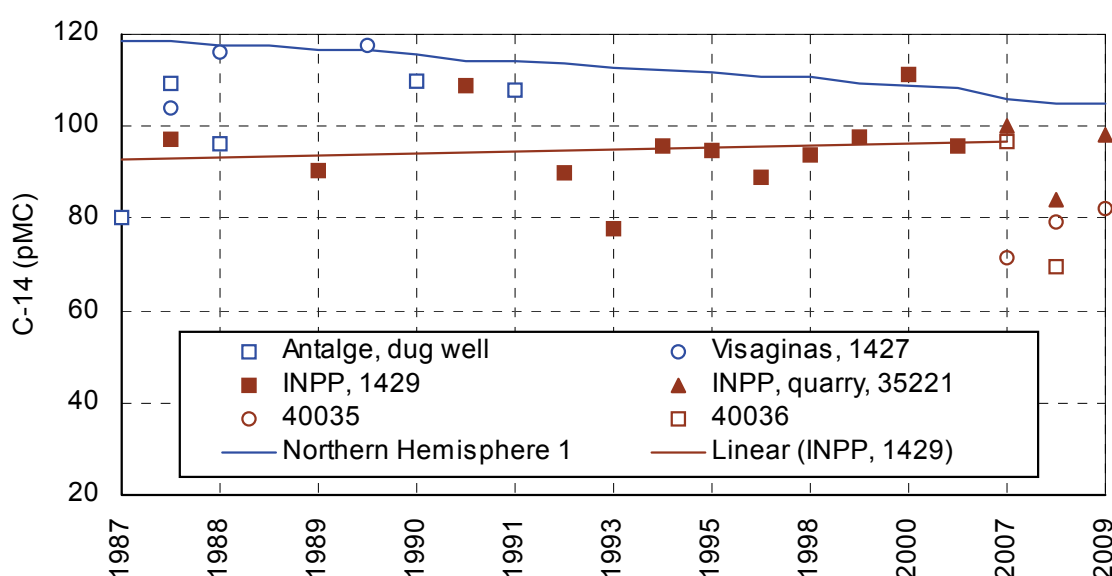

Fig. 8. The ^{14}C specific activity of DIC in groundwater at short distance from INPP for the period 1987–2009 (Antalge, shallow dug well, and Visaginas, well No 1427, are located at long distance from INPP)

As regards the ^{14}C data in pMC units, we found out that all ^{14}C values in the groundwater are close to or below the atmospheric air ^{14}C background line. ^{14}C in DIC of groundwater from some places is originally diluted by HCO_3^- with low isotope ratio ^{14}C/^{12}C. However the ^{14}C trend line for well No 1429 has a slightly increasing tendency what is in opposite with the atmospheric background trend and traces week INPP impact. As groundwater

carbonate system under different aquifer conditions may have different DIC sources and bicarbonate concentrations, we calculated ^{14}C activity concentration from two experimentally measured values such as HCO_3^- concentration and ^{14}C specific activity relative to carbon (Fig. 9).
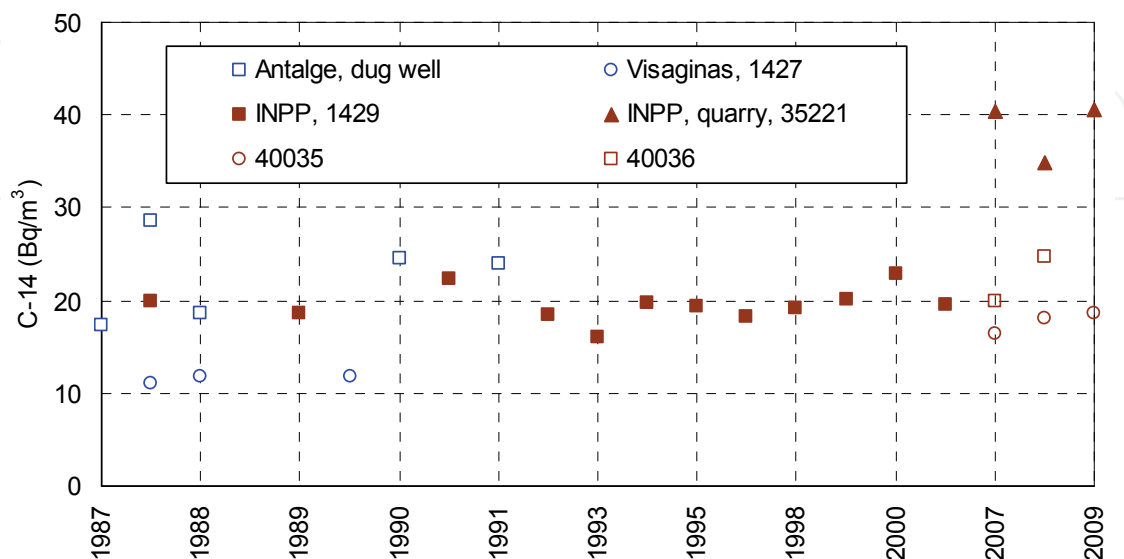

Fig. 9. The ^{14}C activity concentration in groundwater at short distance from INPP for the period 1987–2009 (Antalge, shallow dug well, and Visaginas, well No 1427, are located at long distance from INPP)

In this case, the influence of INPP on ^{14}C content in the groundwater was possible to notice for one quarry observation well. In 2007–2009, the ^{14}C activity concentration in the groundwater from observation well 35221 of the quarry was higher (40.4±0.4 Bq/m³) than the activity in other observed wells (10-20 Bq/m³) due to high HCO_3^- concentration (up to 930 mg/l). In the quarry, the sludge with high concentration of organics from the Visaginas wastewater treatment plant was disposed of and covered with an artificial soil layer. The sludge organics contained enhanced levels of ^{14}C and other radionuclides, firstly ^{60}Co. Due to microbial oxidation of organics under closed conditions, CO_2 generation with enhanced ^{14}C content might have taken place what formed high HCO_3^- concentration in the groundwater. Moreover, the activity concentration of ^{60}Co in the water from quarry observation well No 35221 was measurable – up to 11 Bq/m³ while in water from other observation wells ^{60}Co was never detected. As it was evidenced from the long-term ^{14}C data series, the groundwater systems are closed from contamination by radionuclides and are rarely and insignificantly influenced by the INPP.

3.4 Some data on ^{14}C in reactor process media

As the main circulation circuit water (primary circuit water) and some other water streams of INPP are continuously purified, ionic species such as $H^{14}CO_3^-$ will be adsorbed onto the ion-exchange resins and concentrated. After use, the spent ion-exchange resins will be finally cemented and disposed of as LILW in NSR. The activity of resins comprises the main

fraction of radioactive inventory in the various waste forms disposed of in LILW facilities (Magnusson, 2007).

Different types of samples from the INPP process media were preliminary considered for determination of ^{14}C total specific activity (sum of organic and inorganic species): water from the spent nuclear fuel pools, water from the primary circuit, evaporator concentrates, and spent ion exchange resins (Table 3).

Sample No	Sample description and taking date	[14]C activity concentration
1	Water from the primary circuit, 22 July 2002	<0.05 Bq/l
2		0.10 ± 0.04 Bq/l
3		<0.05 Bq/l
4		<0.05 Bq/l
5	Water from the spent nuclear fuel pools, 12 July 2002	0.64 ± 0.08 Bq/l
6		0.70 ± 0.09 Bq/l
7		0.25 ± 0.04 Bq/l
8	Evaporator concentrates, July 2002	0.55 ± 0.12 MBq/l
9	Spent ion exchange resins, November 2002	70 ± 9 kBq/kg

Table 3. [14]C in process water and operational radioactive waste of INPP

The averaged specific activity of ^{14}C in water samples was estimated as follows: from the primary circuit - less than detection limit (0.05 Bq/l); from the spent nuclear fuel pools - 0.5 Bq/l. Considerably higher specific activity of ^{14}C was determined in evaporator concentrates and spent ion exchange resins - 0.55 MBq/l and 70 kBq/kg, respectively.

Low ^{14}C activity in reactor water of BWRs was reported in (Magnusson, 2007) as well. As in case with BWRs, the low ^{14}C activity in RBMK reactor water illustrates effective purification of water from inorganic ^{14}C species by means of ion exchange resins. Other results on ^{14}C in the INPP process media are in line with ^{14}C data for BWRs presented in (Magnusson, 2007). The obtained data were used for the calculation ^{14}C/^{60}Co scaling factors applied to the RBMK–1500 reactor waste characterization.

4. Conclusion

The ^{14}C concentration near the INPP has been studied by analysing various samples from terrestrial and aquatic environments and groundwater using conventional liquid scintillation counting. A comparison between the specific activity of ^{14}C at short distances from the INPP and ^{14}C specific activity of the background level has revealed the ^{14}C excess over the whole operation period of the INPP (the end of 1983–the end of 2009).

By studying tree rings and annual terrestrial plants in the terrestrial environment mostly continuous ^{14}C excess was delineated recently approaching 10 pMC that can be formed by [14]CO_2 annual release of magnitude 10^{13} Bq with the maximal value of normalized release rate 11 TBq×GW(e)$^{-1}$×year^{-1}. In comparison with the other radionuclide releases from the INPP 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. Nevertheless, this dose estimate is much lower than the ^{14}C dose occurring from the ^{14}C of natural production in the atmosphere (~$12×10^{-3}$ mSv/year).

In the aquatic environment, the ^{14}C activity in DIC from the cooling basin increased only in 2002–2006 with the ^{14}C excess of 30–35 pMC above the background level what could be caused by the ^{14}C release rate from the INPP to water $\sim 10^8$ Bq/year. The highest ^{14}C excess in aquatic plants (up to 65 pMC) with significant variability was detected in the period 1995–2008. The groundwater system was rarely and insignificantly influenced by the INPP. Thus the effective dose resulting from the ^{14}C in the aquatic environment was very low.

It was confirmed that the majority of ^{14}C contained in the gaseous waste (e.g. reactor off-gases) was released from the INPP to the atmosphere during the operation time with the total ^{14}C activity roughly of $\sim 2.0 \times 10^{14}$ Bq. Only a small quantity of ^{14}C was released via liquid effluents from the INPP. A large quantity of ^{14}C is retained in the structural materials of the reactor (graphite moderator mainly with ^{14}C of 7.82×10^{14} Bq per two units) and in the radioactive waste (LILW with ^{14}C of 1.43×10^{13} Bq), which will be handled during the INPP decommissioning phase. For waste containing high levels of ^{14}C, such as graphite, that may exceed the waste acceptance requirements for the near surface disposal, no disposal facility is available at present. This waste only can be kept in an interim storage until a suitable disposal facility (e.g. deep geological disposal) or other disposal and treatment alternatives become available.

5. References

Almenas, K.; Kaliatka, A. & Uspuras, E. (1998). *Ignalina RBMK-1500, a Source Book*, Lithuanian Energy Institute, ISBN 9986-492-35-1, Kaunas, Lithuania

Ancius, D.; Ridikas, D.; Remeikis, V.; Plukis, A.; Plukiene, R. & Cometto, M. (2004) Evaluation of the activity of irradiated graphite in the Ignalina Nuclear Power Plant RBMK-1500 reactor. *Nukleonika*, Vol. 50, No. 3, 113-120, ISSN 0029-5922

Arslanov, Kh.A. (1985). *Radiocarbon: Geochemistry and Geochronology*, Leningrad University Press, Leningrad (in Russian)

Buchuev, A.V.; Zubarev, V.N. & Prochin, I.M. (2002). Composition and quantity of impurities in graphite of industrial reactors. *Atomnaya energija*, Vol. 92, No. 4. 298-302, ISSN 0004-7163 (in Russian)

Buzinny, M.; Kovalyukh, N.; Likhtarjov, I.; Los, I.; Nesvetajlo, V.; Pazdur, M.F.; Skripkin, V.; Shkvorets, O. & Sobotovich, E. (1995). Ecological chronology of nuclear fuel cycle sites. *Radiocarbon*, Vol. 37, No. 2, 469-473

Clark, I.D. & Fritz, P. (1997). *Environmental isotopes in hydrogeology*, Lewis, Boca Raton, FL

Fairhall, A.W. & Young, J.A. (1970). *Radionuclides in the environment*, Advances in Chemistry Series Vol. 93., American Chemical Society, Washington, D.C.

Gaiko, V.B.; Korablev, N.A.; Solovev, E.N.; Trosheva, T.I.; Shamov, V.P.; Umanets, M.P. & Shcherbina, V.G. (1985). Discharge of carbon-14 at nuclear power plants with RBMK-1000. *Atomnaya energija*, Vol. 59, No. 2, 144-145, ISSN 0004-7163 (in Russian)

Gupta, S.H. & Polach, H.A. (1985). *Radiocarbon Practices at ANU – Handbook*, ANU, Canberra

Gusev, N.G. & Beliajev, V.A. (1986). *Radioactive releases into biosphere, reference book*, Energoatomizdat, Moscow (in Russian)

Hertelendi, E. & Csongor, E. (1982). Antrophogenic ^{14}C excess in the troposphere between 1951 and 1978 measured in tree rings. *Radiochemical and Radioanalytical Letters*, Vol. 56, No. 2, 103-110

http://www.iae.lt

Hua, Q. & Barbetti, M. (2004). Review of tropospheric bomb ^{14}C data for carbon cycle modeling and age calibration purposes. *Radiocarbon*, Vol. 46, 1273-1298

IAEA (2004). *Management of waste containing tritium and carbon-14*, Technical Report Series No. 421, International Atomic Energy Agency, Vienna, Austria

IAEA (2006). *Characterization, treatment and conditioning of radioactive graphite from decommissioning of nuclear reactors*, IAEA-TECDOC-1521, International Atomic Energy Agency, Vienna, Austria

Kovaliukh, N.N. & Skripkin, V.V. (1994). An universal technology for oxidation of carbon-containing materials for radiocarbon dating, *Abstract and Papers of Conference on Geochronology and Dendrochronology of Old Town's and Radiocarbon Dating of Archaeological Findings*, pp. 37-42, ISBN 9986-19-061-4, Vilnius, October – November 1994, Vilnius University Press, Vilnius, Lithuania

LAND 42–2001. *Limits of discharge of radioactive substances from nuclear facilities and basic requirements for monitoring of radioactive effluents and monitoring of the environment*, Normative document of the Ministry of Environment of the Republic of Lithuania, State Journal, 2001, No. 13-415

Levin, I.; Kromer, B.; Barabas, M. & Münnich, K.O. (1988). Environmental distribution and long-term dispersion of reactor [14]CO_2 around two German nuclear power plants. *Health Physics*, Vol. 54, No. 2, 149-156

Libby, W.F. (1946). Atmospheric helium three and radiocarbon from cosmic radiation. *Physical Review*, Vol. 69, 671-672

Loosli, E.H. & Oeschger, H. (1989). [14]C in the environment of Swiss nuclear installations. *Radiocarbon*, Vol. 31, No. 3, 747-753

Lukauskas, D.; Plukiene, R.; Plukis, A.; Gudelis, A.; Duskesas, G.; Juodis, L.; Druteikiene, R.; Lujaniene, G.; Luksiene, B. & Remeikis, V. (2006). Method to determine the nuclide inventory of low-activity waste of the RBMK-1500 reactor. *Lithuanian Journal of Physics*, Vol. 46, No. 4, 497-503, ISSN 1392-1932

Magnusson, Å. (2007). [14]C Produced by Nuclear Power Reactors – Generation and Characterization of Gaseous, Liquid and Solid Waste, Lund University, Sweden, *Doctoral Dissertation*, ISBN 978-91-628-7248-9

Magnusson, A.; Stenström, K.; Skog, G., Adliene, D.; Adlys, G.; Hellborg, R.; Olariu, A.; Zakaria, M.; Rääf, C. & Mattsson, S. (2004). Levels of ^{14}C in the terrestrial environment in the vicinity of two European nuclear power plants. *Radiocarbon*, Vol. 46, No. 2, 863-868

Mazeika, J. & Motiejunas, S. (2003). Evaluation of aquatic routine radioactive releases from the Ignalina Nuclear Power Plant. *Ekologija*, No. 4, 36-42, ISSN 0235-7224

Mazeika, J. (2002). *Radionuclides in Geoenvironment of Lithuania*, Institute of Geology, ISBN 9986-615-32-1, Vilnius, Lithuania

Mazeika, J.; Petrosius, R. & Pukiene, R. (2008). Carbon-14 in tree rings and other terrestrial samples in the vicinity of Ignalina Nuclear Power Plant, Lithuania. *Journal of Environmental Radioactivity*, Vol. 99, 238-247

Methodology for assessing the radiological consequences of routine releases of radionuclides to the environment (1995), Radiation Protection 72-Report-EUR 15760EN, Brussels-Luxembourg

Mikhajlov, N.D.; Kolkovsky, V. & Pavlova, I.D. (1999). Radiocarbon distribution in northwest Belarus near the Ignalina nuclear power plant. *Radiocarbon*, Vol. 41, No. 1, 75-79

Milton, G.M. & Kramer, S.J. (1998). Using ^{14}C as a tracer of carbon accumulation and turnover in soils. *Radiocarbon*, Vol. 40, No. 2, 999-1011

Milton, G.M.; Kramer, S.J.; Brown, R.M.; Repta, C.J.W.; King, K.J. & Rao, R.R. (1995). Radiocarbon dispersion around Canadian nuclear facilities. *Radiocarbon*, Vol. 37, No. 2, 485-496

Motiejunas, S.; Nedveckaite, T.; Filistovic, V.; Mazeika, J.; Morkeliunas, L. & Maceika, E. (1999). Assessment of environmental impact due to radioactive effluents from Ignalina NPP. *Environmental and Chemical Physics*, Vol. 21, No. 1, 8-18, ISSN 1392-740X

NCRP (1985). *Carbon-14 in the environment*, National Council on Radiation Protection and Measurements, NCRP Report No. 81, USA

Nedveckaite, T.; Motiejunas, S.; Kucinskas, V.; Mazeika, J.; Filistovic, V.; Jusciene, D.; Maceika, E.; Morkeliunas, L. & Hamby, D.M. (2000). Environmental releases of radioactivity and the incidence of thyroid disease at the Ignalina Nuclear Power Plant. *Health Physics*, Vol. 79, No. 6, 666-674

Obelic, B.; Krajcar-Bronic, I.; Srdoc, D. & Horvatinic, N. (1986). Environmental ^{14}C levels around the 632 MWe nuclear power plant KRSKO in Yugoslavia. *Radiocarbon*, Vol. 28, No. 2A, 644-648

Plukis, A.; Remeikis, V.; Juodis, L.; Plukiene, R.; Lukauskas, D. & Gudelis, A. (2008). Analysis of nuclide content in Ignalina NPP. *Lithuanian Journal of Physics*, Vol. 48, No. 4, 375-379

Raaen, F.F.; Ropp, G.A. & Raaen, H.P. (1968). *Carbon-14*, McGraw-Hill, New York

Stenström, K.; Erlandson, B.; Hellborg, R.; Wiebert, A. & Skog, G. (1996). Determination of the $^{14}CO_2$ and the total airborne ^{14}C releases from two Swedish light-water reactors using accelerator mass spectrometry. *Radioactivity and Radiochemistry*, Vol. 7, No. 1, 32-36

Stuiver, M. & Polach, H.A. (1977). Reporting of ^{14}C data. *Radiocarbon*, Vol. 19, 355-363

Uchrin, G.; Hertelendi, E.; Volent, G.; Slavik, O.; Morávek, O.; Kobal, I. & Vokal, B. (1998). Carbon-14 measurements at PWR type nuclear power plants in three Middle European countries. *Radiocarbon*, Vol. 40, No. 1, 439-447

Veres, M.; Hertelendi, E.; Uchrin, G.; Csaba, E.; Barnabás, I.; Ormai, P.; Volent, G. & Futó, I. (1995). Concentration of radiocarbon and its chemical forms in gaseous effluents, environmental air, nuclear waste and primary water of a pressurized water reactor power plant in Hungary. *Radiocarbon*, Vol. 37, No. 2, 473-497
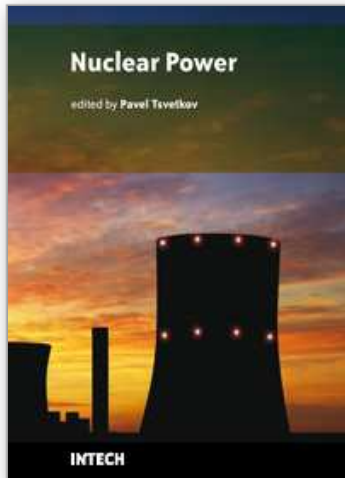
Nuclear Power

Edited by Pavel Tsvetkov

The world of the twenty first century is an energy consuming society. Due to increasing population and living standards, each year the world requires more energy and new efficient systems for delivering it. Furthermore, the new systems must be inherently safe and environmentally benign. These realities of today's world are among the reasons that lead to serious interest in deploying nuclear power as a sustainable energy source. Today's nuclear reactors are safe and highly efficient energy systems that offer electricity and a multitude of co-generation energy products ranging from potable water to heat for industrial applications. The goal of the book is to show the current state-of-the-art in the covered technical areas as well as to demonstrate how general engineering principles and methods can be applied to nuclear power systems.

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