

LEVELS OF ^{14}C IN THE TERRESTRIAL ENVIRONMENT IN THE VICINITY OF TWO EUROPEAN NUCLEAR POWER PLANTS

Åsa Magnusson[1] • Kristina Stenström[1] • Göran Skog[2] • Diana Adliene[3] • Gediminas Adlys[3] • Ragnar Hellborg[1] • Agata Olariu[4] • Mohamad Zakaria[5] • Christopher Rääf[5] • Sören Mattsson[5]

ABSTRACT. Radiocarbon is produced in all types of nuclear reactors. Most of the ^{14}C released into the environment is in the form of gaseous emissions. Recent data on the ^{14}C concentration found in terrestrial samples taken in the vicinity of nuclear power plants in Romania and Lithuania are presented. We found increased ^{14}C levels in the surroundings of both power plants. At the Romanian power plant Cernavoda, we found excess levels of ^{14}C in grass within a distance of about 1000 m, the highest ^{14}C specific activity being 311 Bq/kg C (approximately 28% above the contemporary ^{14}C background) found at a distance of 200 m from the point of release (nearest sampling location). At the Lithuanian power plant Ignalina, samples of willow, pine, and spruce showed a ^{14}C excess of similar magnitude, while significantly higher values were found in moss samples. The samples were analyzed at the accelerator mass spectrometry facility in Lund, Sweden.

INTRODUCTION

Radiocarbon is produced in all types of reactors through neutron-induced reactions with isotopes of carbon, nitrogen, and oxygen present in the reactors. Since most of the ^{14}C released into the environment is in the form of gaseous emissions (such as $^{14}CO_2$), terrestrial samples will constitute the primary indicators of increased ^{14}C levels in the surroundings of the nuclear power plant (NPP). Because the food chain starts with plants, measurements of ^{14}C in environmental samples are important for estimates of radiation exposure to the public.

We have investigated the ^{14}C levels in the surroundings of 2 different types of reactors: the Romanian Canadian Deuterium Uranium (CANDU) reactor at Cernavoda and the Lithuanian RBMK (light-water-cooled, graphite-moderated) reactor at Ignalina. These 2 types of reactors are known to release higher amounts of airborne ^{14}C than light-water-moderated reactors. According to UNSCEAR (2000), the mean normalized ^{14}C release to air from 1990–1994 from CANDU reactors was 1.6 $TBq/GW_e{\cdot}yr$, and for RBMK reactors, 1.3 $TBq/GW_e{\cdot}yr$ (estimated). Corresponding values for light-water-moderated pressurized water reactors (PWRs) and boiling water reactors (BWRs) were 0.22 and 0.51 $TBq/GW_e{\cdot}yr$, respectively. Published data involving ^{14}C levels in the areas surrounding the Cernavoda and Ignalina NPPs are scarce, which justifies further investigations.

We have collected various vegetation samples in the area surrounding the 2 NPPs. At Cernavoda, we collected grass samples at various distances from the NPP, and from the immediate vicinity of the Ignalina NPP, we collected various vegetation samples.

In this paper, we have included a brief description of the 2 reactor types, as well as a summary of previous measurements involving ^{14}C emission and ^{14}C levels in the area surrounding the 2 NPPs.

[1]Department of Physics, Division of Nuclear Physics, Lund University, Box 118, SE-221 00 Lund, Sweden. Corresponding author. Email: asa.magnusson@nuclear.lu.se.
[2]Department of Geology, Quaternary Geology, Lund University, Sölvegatan 12, SE-223 62 Lund, Sweden.
[3]Department of Physics, Kaunas Technological University, LT-3031 Kaunas, Lithuania.
[4]National Institute for Physics and Nuclear Engineering, RO-76900 Bucharest, Romania.
[5]Department of Radiation Physics, Lund University, Malmö University Hospital, SE-205 02 Malmö, Sweden.

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SITE DESCRIPTIONS AND PREVIOUS MEASUREMENTS

Cernavoda, Romania

The Cernavoda NPP is located 180 km east of Bucharest and is designed for 5 CANDU-6 (600 MW_e) reactors. So far, only 1 reactor has been built—with a maximum electrical output of 706 MW_e—and this has been in operation since April 1996; a second reactor is under construction. The CANDU-6 reactor is heavy-water-moderated and -cooled and uses natural uranium as fuel. The height of the venting stack is 50 m. About 220,000 people live within 30 km of the site (AECL 2001).

The largest contributor (>95%) to the production of ^{14}C in CANDU reactors is neutron activation of ^{17}O in the heavy-water moderator. Minor quantities are generated in the heat transport system by the annulus gas (CO_2) system (IAEA 2002) and in the fuel elements (Milton 1995).

According to Boss and Allsop (1995) and ACRP (1995), the total ^{14}C production in a CANDU-6 reactor is about 18 TBq/yr (approximately 33 $TBq/GW_e{\cdot}yr$, assuming an electrical output of 0.600 GW_e and operation of 90% of the time) and 28 $TBq/GW_e{\cdot}yr$, respectively, and it has been estimated that less than 4% is released to the atmosphere (ACRP 1995). Measurements have shown that about 70% of this ^{14}C is released in the form of CO_2 (Cooper 1998).

According to Dubourg (1998), the approximate release rate of ^{14}C with gaseous effluents from a CANDU-6 reactor (assuming operation of 80% of the time) is 4.1 TBq/yr (approximately 8.5 $TBq/GW_e{\cdot}yr$, assuming an electrical output of 0.600 GW_e and 80% operation). Baciu et al. (1996) report that "the normal operation source term of ^{14}C in gaseous effluents" from the CANDU-6 at Cernavoda is 5.7 TBq/yr (approximately 11 $TBq/GW_e{\cdot}yr$, assuming an electrical output of 0.600 GW_e and 90% operation). However, since these data were reported before the NPP was operational, the value probably refers to typical values for CANDU-6 reactors.

Bobric and Simionov (1999) presented annual ^{14}C emission data from the Cernavoda NPP, measured at the release point, from April 1996 to December 1998. The values are: 0.035 TBq/yr (0.26 $TBq/GW_e{\cdot}yr$), 0.18 TBq/yr (0.32 $TBq/GW_e{\cdot}yr$) and 0.29 TBq/yr (0.52 $TBq/GW_e{\cdot}yr$) for 1996, 1997, and 1998, respectively. They also measured ^{14}C concentrations in terrestrial biota (e.g. vegetables and fruits) within 30 km of the NPP but did not find any detectable amounts with the liquid scintillation counter used.

Tenu et al. (2002) measured ^{14}C in atmospheric CO_2 samples collected monthly from April 1995 to November 1998 at a site close to the Cernavoda NPP. During this period, ^{14}C activities corresponding to values ranging from 210–440 Bq/kg C were recorded, with large fluctuations. Increased ^{14}C levels, compared with the values from the previous year, were found during the same month as the NPP started (April 1996). During 1998, they recorded an increase in ^{14}C activity at the Cernavoda site, from a level corresponding to 250 Bq/kg C in April 1998 to 440 Bq/kg C a month later. The values remained above 280 Bq/kg C until the end of the year.

Ignalina, Lithuania

The Ignalina NPP is located close to the border between Belarus and Latvia and consists of 2 units which were put into operation in 1983 and 1987, respectively. The reactors are graphite-moderated BWRs of channel type (model RBMK-1500) and have a maximum electrical output of 1300 MW_e each. The height of the venting stack is 150 m. About 210,000 inhabitants live within 30 km of the power plant (Almenas et al. 1998).

The emission of ^{14}C from a RBMK-1500 reactor mainly originates from ^{14}C production in the nitrogen-helium mixture that fills the entire reactor space and from ^{14}C production in the coolant. During the initial period of reactor operation, pure nitrogen is used instead of the nitrogen-helium mixture. During this period (approximately 18 months), the formation of ^{14}C is about 10 times higher than during the subsequent normal operation at rated power (Konstantinov 1989). Measurements performed in 1985–1986 of atmospheric ^{14}C release at the high-altitude pipe gave a value of 1.4 ± 0.3 TBq/GW_e·yr (Konstantinov 1989).

Mikhajlov et al. (1999) measured the ^{14}C distribution near the Ignalina NPP, and the highest value found corresponds to a ^{14}C specific activity of 430 Bq/kg C recorded in 2-yr-old pine cones that were collected at a distance of 5 km east of the location. Jakimaviciute-Maseliene et al. (2003) measured the ^{14}C concentration in plants (*Artemisia L.* [stem] and *Alnus L.* [leaves]) in the surroundings of the Ignalina NPP in 1996 and 2001. The highest value they found, within 1 km northwest of the NPP, corresponds to a ^{14}C activity of 400 Bq/kg C (in *Alnus L.* in 2001). From this maximum value, they derived a rough value of the atmospheric ^{14}C release from the Ignalina NPP of 49 TBq/yr (approximately 38 TBq/GW_e·yr, derived from the 2 units mean electrical energy generated during 1996 and 2001).

SAMPLING

Grass samples were collected in 1999 (14 October) and in 2001 (7 September) at various distances (200–4300 m) from the Cernavoda NPP. On the first sampling occasion, 9 samples were collected for ^{14}C analysis, and on the second, 21 samples were collected. The sampling locations and the general wind direction at Cernavoda are shown in Figure 1.

Different types of vegetation samples from trees and moss were collected in the immediate vicinity of the Ignalina NPP on 10 April 2003. All 9 samples were taken within a distance of 400 m from the stack. The approximate sampling locations and the general wind direction at Ignalina can be seen in Figure 2. Additional details are given in Table 1.

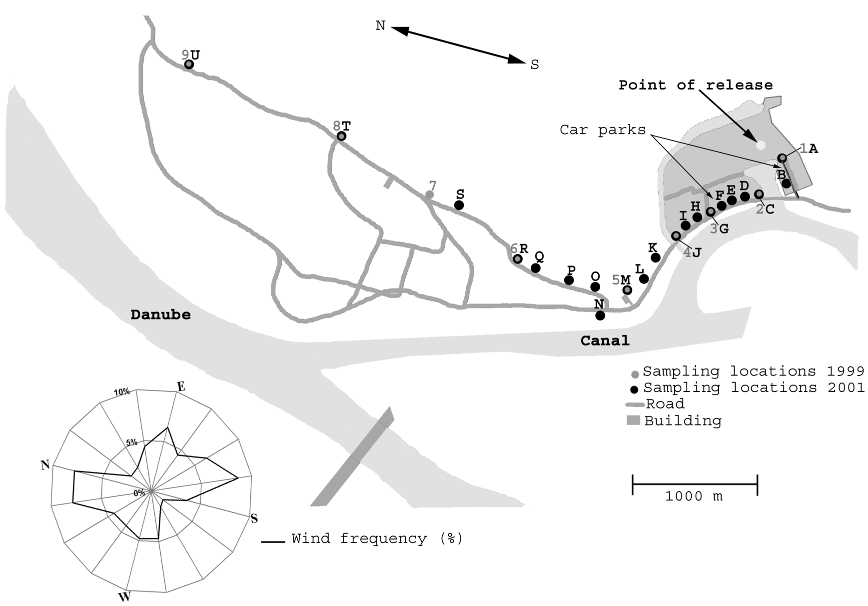

Figure 1 Sampling locations at Cernavoda in 1999 and 2001 and the wind frequency at Cernavoda from March to October 1999.

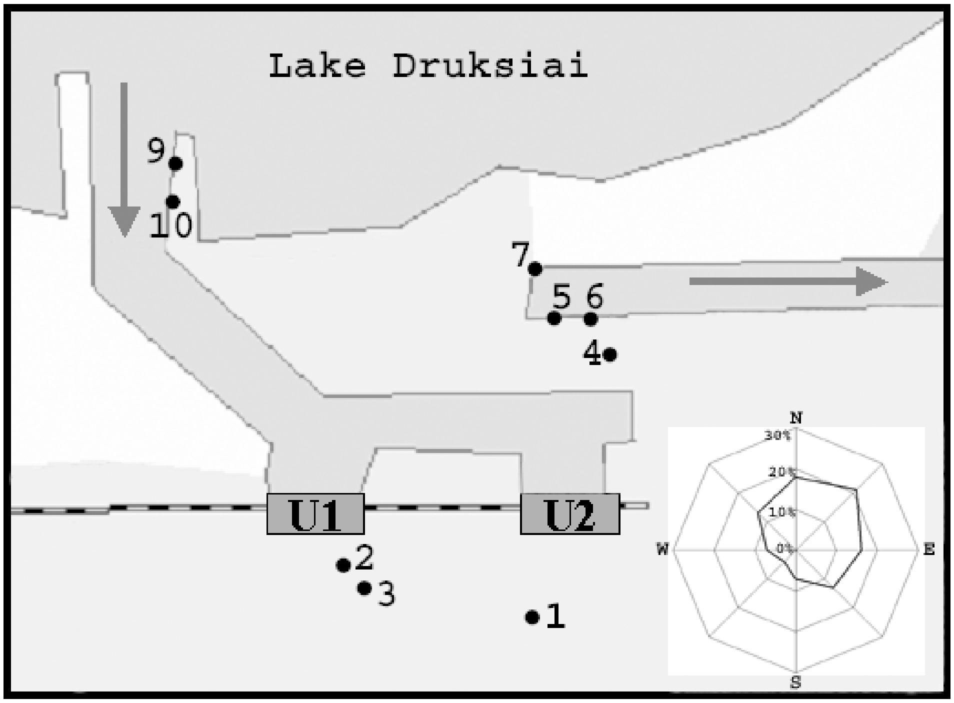

Figure 2 Approximate sampling locations at the Ignalina NPP in April 2003 (additional information is given in Table 1) showing Unit 1 and 2. The wind frequency at Ignalina during 2002 is also shown.

The samples were dried, ground, combusted, and graphitized according to standard procedures at our laboratory (Stenström 1995) and analyzed at the accelerator mass spectrometry facility in Lund, Sweden. The moss samples were fractionated into 1 upper layer (mainly moss) and 1 or 2 lower layers (moss and soil).

RESULTS

Cernavoda, Romania

The results from Cernavoda, which can be seen in Figure 3, show the typical distribution of ^{14}C from a point source. Excess levels of ^{14}C were found within a distance of about 1000 m from the point of release, with a maximum ^{14}C specific activity of 311 Bq/kg C found at the closest sampling location (A, 200 m).

Ignalina, Lithuania

The data given in Table 1 from the Ignalina NPP show similar levels of ^{14}C in willow, pine, and spruce to those found in grass from Cernavoda. The concentration in the moss samples and in the soil is significantly higher.

DISCUSSION

Increased ^{14}C levels were found in the surroundings of both NPPs. At Cernavoda, excess levels of ^{14}C were found in grass up to a distance of about 1000 m. Similar values of ^{14}C excess were found in the samples of leaves, shoots, and twigs from trees and bushes close to the Ignalina NPP. These do not necessarily reflect the maximum levels since they were collected only in the immediate

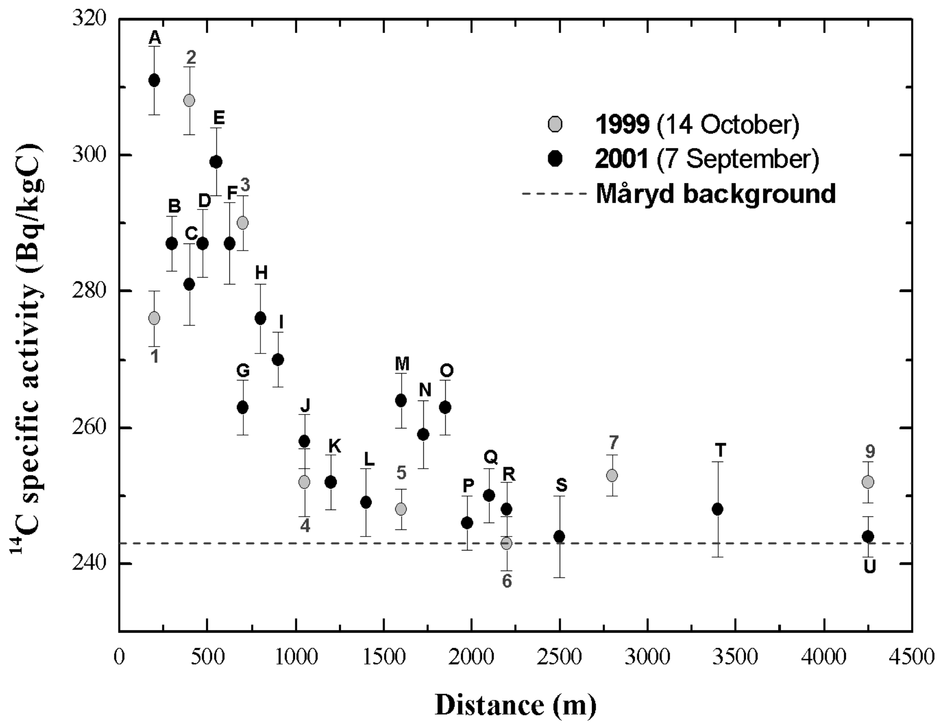

Figure 3 The ^{14}C specific activity (Bq/kg C) found in grass samples collected at various distances from the Cernavoda NPP. The background ^{14}C activity, 243 ± 2 Bq/kg C, measured in rush (*Juncus L*) from Måryd, Sweden, is the mean value for the period 1999–2001. Figures and numbers refer to the sampling times and locations shown in Figure 1.

Table 1 ^{14}C specific activity (Bq/kg C) found in various vegetation samples collected in the immediate vicinity of the Ignalina NPP in April 2003. The sampling locations are indicated in Figure 2. U1 and U2 = Units 1 and 2. The background ^{14}C activity, 241 ± 2 Bq/kg C, measured in rush (*Juncus L.*) from Måryd, Sweden, is the mean value for the period 2002–2003.

Nr	Sample description	Location	Background activity (Bq/kg C)
1	Goat willow (shoot, twig and flower bud)	150 m in front of U2	302 ± 7
2	Pine (shoot and twig)	50–70 m in front of U1	315 ± 7
3	Spruce (shoot and twig)	100 m in front of U1	302 ± 10
4	Goat willow (shoot and twig)	200 m behind U2	309 ± 7
5a	Moss (upper layer of a few cm)	Along outlet canal	362 ± 6
5b	Moss (soil and moss from lower layer)		1960 ± 20
6a	Moss (upper layer of a few cm)	Along outlet canal	370 ± 7
6b	Moss (soil from lower layer)		598 ± 10
7	White willow (shoot and twig)	Beginning of outlet canal	323 ± 7
9	Goat willow (shoot and twig)	Along inlet canal	342 ± 5
10a	Moss (upper layer of a few cm)	Along inlet canal	1020 ± 10
10b	Moss (middle layer of a few cm)		1570 ± 20
10c	Moss (soil and moss from lower layer)		4730 ± 60

vicinity (within 400 m) of the NPP. Because of the considerable height of the venting stack, the maximum ^{14}C concentrations are most likely found at a distance beyond the 400 m. High ^{14}C specific activities were found in the moss samples from Ignalina, with increasing concentrations with increasing depth. As moss grows from the top, the different layers should reflect different time periods, the oldest being the bottom layer. The results obtained may thus indicate that the release of ^{14}C has decreased during the lifetime of the moss. But as the ^{14}C concentrations found in the moss samples are higher than expected, the possibility of airborne ^{14}C particulates must be considered (Marsden et al. 2002; Mikhajlov et al. 1999). According to UNSCEAR (2000), the amount of particulates released with airborne effluents from the Ignalina NPP is considerable. Because of the great number of factors that determine depletion of particulates from a plume, it is extremely difficult to relate the concentration of assumed ^{14}C particulates found on the ground to the emission rate of these particulates.

Further data on moss samples and soil profiles at larger distances (up to ~30 km) from Ignalina NPP will be reported. This study was partly financed by grants from the Swedish Radiation Protection Authority and Carl Tryggers Stiftelse.

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