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MEASUREMENTS OF ^{14}C IN TREE-RINGS AROUND THE KRŠKO NUCLEAR POWER PLANT

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ABSTRACT

Radiocarbon, ^{14}C, artificially produced in nuclear fuel cycle facilities, is one of the most important global radioactive contaminants. Nuclear power plant emissions of ^{14}C to the environment are mainly via the stack gas. The dominating chemical forms of ^{14}C released from PWR reactors are hydrocarbons and to a lesser extent carbon-dioxide. ^{14}C emitted in the form of carbon-dioxide incorporates into plants by photosynthesis. The objective of our work was to determine ^{14}C activity in tree-rings, dated from the period 1975 to 1994, in the surroundings of the 632 MWe Krško Nuclear Power Plant, which began operation in 1981.

Three acacia trees (*Robinia sp.*) within a radius of a few kilometres from the power plant stack at different locations in the prevailing wind direction were felled and individual tree-rings were separated. Samples were first combusted to CO_2, which then reacted with hot lithium to form Li_2C_2. Addition of tritium-free water to the cooled lithium carbide produced acetylene, then the acetylene was catalytically trimerized to benzene. A scintillator (butyl-PBD) was dissolved in benzene and then the samples were counted by a low level liquid scintillation counter (Quantulus LKB-Wallac).

The results of our measurements are presented as ^{14}C activity as a percentage of the modern standard (0.95 of NBS oxalic acid). No influence of power plant releases was observed in the tree-ring ^{14}C activities, which is in accordance with previous measurements of ^{14}C activity in tree-rings of a linden tree felled at Libna, 1.5 km from the power plant.

INTRODUCTION

Radiocarbon, ^{14}C, is naturally created in the upper layers of atmosphere from the capture of cosmic ray produced neutrons (n) by nitrogen (^{14}N) resulting in the emission of protons (p) and the creation of ^{14}C atoms [$^{14}N(n,p)^{14}C$]. ^{14}C is then oxidized to $^{14}CO_2$ and distributed evenly throughout the global atmosphere (Gupta and Polach, 1985). Plants assimilate ^{14}C during photosynthesis, and animals eat plants. The same occurs in the sea, because $^{14}CO_2$, like normal CO_2, dissolves in the oceans and is available to plankton, coral, mollusc and fish. So all organisms during their life continuously replenish their ^{14}C content. At the same time ^{14}C decays following the law of radioactivity with a half life of 5730 years. These two processes lead to an equilibrium of the radiocarbon content in the atmosphere and biosphere.

After the death of plants or animals ^{14}C input ceases. The time elapsed from death can be established by determining the residual ^{14}C content. Changes in atmospheric $^{14}CO_2$ abundance are particularly important as all life forms, irrespective of their location, derive their ^{14}C primarily from atmospheric $^{14}CO_2$.

^{14}C is artificially produced in nuclear fuel facilities and is one of the most important radionuclides released to the environment from nuclear power plants (Uchrin et al., 1993). It is produced primarily (i) in the fuel by the reactions $^{17}O(n,\alpha)^{14}C$ and $^{14}N(n,p)^{14}C$, (ii) in core structural materials, including stainless steel, where the source of ^{14}C is nitrogen

and (iii) in the cooling water where the sources of ^{14}C are oxygen and nitrogen containing substances. ^{14}C is released to the environment mainly via the stack. ^{14}C discharges by liquid and solid wastes are less than 5% of the gaseous release. The dominating chemical forms of radiocarbon released from BWR type reactors are carbon dioxide and to a lesser extent hydrocarbons, while PWR type of reactors emit radiocarbon mostly in the form of hydrocarbons (Hertelendi et al., 1989; Uchrin et al., 1992).

In our work ^{14}C activity was determined in tree-rings in trees growing around the 632MW Krško Nuclear Power Plant, which began operation in 1981.

SAMPLING POINTS

Krško Nuclear Power Plant is located in the south-east part of Slovenia, on the left bank of the River Sava in the industrial zone of Krško (45° 45' 15" N, 15° 31' 14" E). The immediate and surrounding area of the power plant belongs partly to the wooded secondary mountain ranges of the Alps to the north and to the wooded secondary mountain ranges of the Dinaric Mountains to the west. The major part belongs to the wine-growing Panonian Boundary Hills and the Panonian Plain. The climate is moderate and partly influenced by the nearby Alps. The humidity is relatively high (85%), with frequent fog, especially in the River Sava basin. The region is known for moderate winds, mainly along the River Sava flow (NW-SE).

To reconstruct past activity of ^{14}C in the region, three acacia trees (*Robinia sp.*) were cut down in the radius of a few kilometres from the power plant stack at two different locations in the prevailing wind direction, as seen from Figure 1. The period covered was from 1975 to 1994.

Figure 1.: Sampling points

METHOD

Samples were first combusted to CO_2. The sample for $\delta^{13}C$ mass spectrometric measurements is collected at this stage. CO_2 was then reacted with hot lithium to form Li_2C_2. Addition of tritium-free water to the cooled lithium carbide produced acetylene, and then the acetylene was catalytically trimerized to benzene. A scintillator compound (butyl-PBD) was dissolved in benzene and the solution was then counted in a low level liquid scintillator counter (Quantulus LKB-Wallac) for 1000 minutes, 10 times for each sample.

RESULTS AND DISCUSSION

The results of the measurements are presented in Table 1 showing the ^{14}C activity as a percentage of modern standard (pM), which is 0.95 of NBS oxalic acid, and $\delta^{13}C$ values which, as expected, are around -25‰. Activities of tree-rings show no significant variations. They agree, within the error bars, with the activity of the atmospheric CO_2 in the Northern Hemisphere.

Table 1.: ^{14}C activity and $\delta^{13}C$ values in tree-rings

	SAMPLE 1		SAMPLE 2		SAMPLE 3	
year	pM	$\delta^{13}C$ (‰)	pM	$\delta^{13}C$ (‰)	pM	$\delta^{13}C$ (‰)
1975			132.0 ± 7.7	-25.66*		
1976			136.4 ± 7.9	-25.81		
1977			135.1 ± 7.8	-25.95		
1978			125.2 ± 7.3	-25.72		
1979	125.3 ± 7.3	-26.16	123.0 ± 7.1	-25.94		
1980			128.8 ± 7.5	-25.92		
1981			122.2 ± 7.1	-25.72		
1982			120.9 ± 7.0	-25.86		
1983			123.0 ± 7.1	-25.92		
1984			118.5 ± 6.9	-25.77	122.5 ±7.1	-26.11
1985					124.5 ± 7.2	-27.05
1986			117.7 ± 6.8	-25.68	120.7 ± 7.0	-27.06
1987					121.4 ± 7.0	-26.04
1988					119.9 ± 7.0	-25.88
1989					121.5 ± 7.1	-25.95
1990			114.0 ± 6.6	-24.64	115.7 ± 6.7	-25.32
1991					119.8 ± 7.0	-25.19
1992					119.2 ± 7.0	-24.72
1993					118.5 ± 6.9	-24.04
1994	110.0 ± 6.4	-25.94	111.6 ± 6.5	-25.40	119.9 ± 7.0	-25.88

* ± 0.05

Figure 2 shows ^{14}C activity in tree-rings (points) as compared with the mean atmospheric ^{14}C activity in the Northern Hemisphere according to Levin et al. (full line) (Levin, et al, 1980)). The results were also compared with the ^{14}C activity of linden tree (*Tilia sp.*),which was felled at Libna, approximately 1.5 km from the plant stack (Obelić, et al, 1986). A sampling site at the Plitvice National Park in central Croatia was chosen as a ^{14}C reference point not affected by the Power Plant. There was no influence of power plant releases observed in tree-ring ^{14}C activities, which is in accordance with the previous measurements of ^{14}C activity in tree-rings of the linden tree from Libna, and with measurements of ^{14}C activity in tree-rings at the Plitvice National Park in central Croatia.

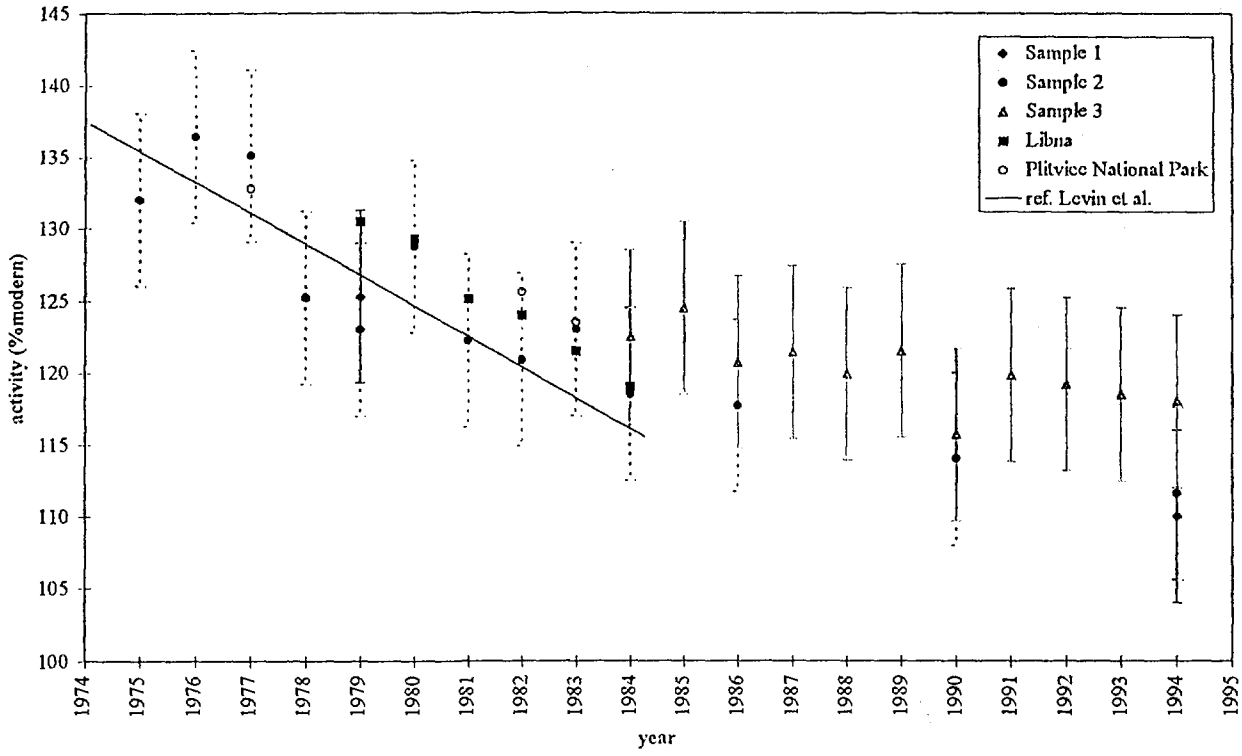

Figure 2.: ^{14}C activity (pM) of acacia tree-rings, 1975-1994 compared with ^{14}C activity of linden tree-rings from Libna and the mean ^{14}C activity in the Northern Hemisphere according to Levin et al. (full line) (Radiocarbon 22, 1980)

CONCLUSION

Our analysis shows that tree-rings follow the general activity trend in the Northern Hemisphere and no enhanced activity due to the power plant release was observed in tree-rings.

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