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TRACES OF ^{14}C EMISSIONS FOR THE OPERATION PERIOD OF TWO UKRAINIAN NPPS: RIVNE AND CHORNOBYL

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ABSTRACT. The aim of this study was a comparative retrospective assessment of radiocarbon (^{14}C) as a tracer, caused by operational emissions of Rivne and Chornobyl nuclear power plants (NPPs), which are equipped with different types of nuclear reactors. For this purpose, ^{14}C was studied in annual tree rings of pine taken at a distance of 1.5 km southwest of the Rivne NPP and at a distance of 3.5 km west-northwest of the Chornobyl NPP, near the Yaniv railway station. As a background, we use the ^{14}C in air data (Hua et al. 2013), which we continue for time interval 2009–2020 with our experimental data for pine tree rings. Tree rings were also collected in a rural area 60 km west of Kyiv, where industrial impact, in our opinion, is absent. ^{14}C in wood samples was determined using the conventional method based on liquid scintillation counting. It was found that the ^{14}C excess in the annual tree-ring samples of pine near the Chornobyl NPP during the observed operation period (1984–2000) was 3.0–13.0 pMC, except for the 1986, the year of the Chornobyl accident, when the ^{14}C value rose sharply to 182.7 pMC (^{14}C excess 62 pMC). After 2000, the content of ^{14}C in the air near the Chornobyl nuclear power plant did not exceed the background values within the uncertainty of the measured data. The concentration of ^{14}C in the samples of annual tree rings of pine near the Rivne NPP for the observation period (1986–2019) corresponded to the background levels within the uncertainty of the measured data. The study of environmental traces of ^{14}C emissions from two NPPs equipped with different types of reactors showed significantly lower emissions of Rivne NPP with VVER compared with emissions from Chornobyl NPP with RBMK reactors.

KEYWORDS: annual tree rings, ^{14}C, NPP, nuclear power plant, operational emissions, traces in the environment.

INTRODUCTION

At the present time, nuclear power plants (NPPs), together with spent nuclear fuel reprocessing plants and radioactive waste repositories, form the main industrial sources of radiocarbon (^{14}C) release into the atmosphere. Areas of maximum ^{14}C excess are located around NPPs at a distance of 1–2 km from the ventilation pipe, however, sometimes could be traced up to a 20–30 km distance (Levin and Kromer 2004; Magnusson 2007).

Emissions of ^{14}C in the form of CO_2 and C_nH_m can occur during operation of NPPs. NPPs’ total ^{14}C emissions are characterized by power capacity and the ratio of their forms, which determines the type of used reactors. The $^{14}CO_2$ component of emissions from currently operating VVER reactors at Ukrainian NPPs is low, ranging from a few to 25% of total ^{14}C emissions (IAEA 2004; Rajec et. al. 2011), which makes it difficult to trace in the environment around facilities (Varga et al. 2020). The main form of ^{14}C emissions of graphite reactors of the RBMK type is CO_2, which allows clear retrospective studies near the NPP (Pabedinskas et al. 2019).

The Ukrainian NPP operator is introducing current monitoring of ^{14}C emissions, taking continuous gas samples from the ventilation pipe. As emission to the environment dilutes

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*Corresponding author. Email: michael.buzinny@gmail.com rapidly, ^{14}C excess traced at a limited distance from the emission source, in particular the component corresponding to ^{14}C emissions in the form of CO_2 can be traced in biota samples.

Today, Rivne NPP has four power units (VVER type) with a total installed capacity of 2835 MW. Four RBMK-1000 reactors operated at the Chornobyl (Chernobyl) NPP until 1986, while two of them continued to operate up to shut down in 2000.

Retrospective studies of the ^{14}C excess component in the annual tree rings caused by $^{14}CO_2$ emissions while operation of post-Soviet NPP have been conducted previously: ^{14}C emissions of the Chornobyl NPP operation for the period before 1996 and for few years operation of the Zaporizhzhya NPP (Buzinny et al. 1994; Buzynnyi and Talerko 2000). Those emissions rates were compared with ^{14}C emission chronology data from a nuclear facility located near Tomsk (Russian Federation) (Buzinny et al. 1995) together with Chornobyl NPP's accidental emissions ^{14}C (Buzinny et al. 1997). As for the Tomsk facility (Buzinny et al. 1995), the measured ^{14}C excess in tree rings was combined with an empirical model of ^{14}C spatial distribution shown in McCartney and Scott (1988). For the Chornobyl NPP, modeling of gas transport (Buzynnyi and Talerko 2000) was performed based on existing data of weather conditions and empirical ^{14}C excess measured in annual tree rings for two locations: to the north and east direction to source (Buikov et al. 1992). The annual emission values were in the range of 0.3–3.3 TBq, which for the operation of the Chornobyl NPP until 1996 accumulated to 20 TBq (Buzynnyi and Talerko 2000).

Annual emissions for Tomsk facility were estimated up to 30–45 TBq when total ^{14}C discharges during of operations (Buzinny et al. 1995) accumulated up to 450–620 TBq. Later ^{14}C studies of the annual tree rings of a pine tree situated at another location around the Tomsk facility (Buzynnyi 2020) clearly showed the significant role of distance and direction from the source on the chronological series of emission data and their ratios.

Another ^{14}C distribution in the annual tree rings of pine, which corresponds to the operation of the Chornobyl NPP, was considered (Skripkin et al. 2005) for a site located approximately 3.5 km south-southeast of the NPP.

^{14}C emissions from NPPs are not limited to long-term regular emissions because an emergency situation at an NPP can lead to a significant emission produced in a short time. The accident at the Chornobyl NPP formed a significant, pronounced trace of ^{14}C around the area. Research of this trace includes the gas component (Buzynnyi 1993; Kovalyukh et al. 1994b) and its spatial distribution (Buzinny et al. 1997). Surficial contamination of 1986 herbarium samples caused significant ^{14}C content in them, which indicated the spread of graphite dust (Buzynnyi et al. 1993). The fate of the graphite component of the Chernobyl NPP emergency release is the subject of research in particular (Kovalyukh et al. 1994a, 1997; Skripkin et al. 2005). Studies devoted to distribution of radioactive graphite in the forest ecosystem (forest litter and topsoil) showed extreme contrast of the spatial distribution (Buzinny 2006; Buzynnyi and Skrypkin 2018).

A study of the radiocarbon content of annual wood rings near the Fukushima Dai-ichi NPP accident site showed that the pronounced predominant direction of propagation was southwest, and the maximum excess level of ^{14}C in the wood in 2011 was 31.2 pMC (Chen et al. 2017). At the same time, the extremely low levels compared to the Chernobyl NPP are related to the different principles of operation of the two nuclear plants. An assessment of the impact of emergency emissions from Fukushima Dai-Ichi, both liquid discharges and

Figure 1 Locations of Chornobyl NPP and corresponding sampling sites around Yaniv railway station and Kopachi (Skripkin et al. 2005).

dry deposits on the content of ^{14}C in the water column of the ocean besides ^{3}H and ^{137}Cs, was carried out (Povinec et al. 2017), which made it possible to estimate the corresponding inventories of ^{3}H and ^{137}Cs as well.

METHODS

The annual wood of pine trees (*Pinus sylvestris* L.) was studied retrospectively with the aim to estimate the ^{14}C excess in the air associated with $^{14}CO_2$ emissions of NPPs. Samples of pine wood (1985–2019) were collected in July 2020 (the tree was cut down in the spring) 1.5 km southwest of Rivne NPP. Another studied pine wood material (1984–2021) was collected in October 2021, 3.5 km west-northwest of the Chornobyl NPP (near Yaniv railway station); see Figure 1 where we indicate the Kopachi sampling location (Skripkin et al. 2005).

As a background, we use values of ^{14}C in air for the corresponding period 1985–2009, given by Hua et al. (2013). We continue background data with our experimental data series for pine tree for the time interval 2009–2020 (tree collected in the spring of 2021 in a rural area 60 km west of Kyiv, where industrial impact, in our opinion, is absent).

In all cases, we used a fragment of a tree trunk, about 15–20 cm long with well-defined annual tree rings. The annual tree ring material was separated and ground using a sharp knife to produce wood shavings. The shavings were soaked for 3 to 5 days to wash out the extractives in a 1:1 mixture of ethyl alcohol and ethyl acetate, after which the wood was dried and then charred. Further charcoal was fused with lithium, and then we obtain benzene by a chain of chemical transformations "coal-carbide-acetylene-benzene." Most of the tree ring samples have initially 25–42 g of wood and only few of them about 10–15 g. To prepare a benzene sample, we had used a corresponding set of equipment. For small wood samples, we had used up to 10 g of dry wood shavings and corresponding technology of vacuum pyrolysis (Skripkin and Kovaliukh 1997) to obtain maximum benzene samples. Benzene sample purification includes adding few drops of sulfuric acid, store for few hours, and final sublimation.

The specific activities of ^{14}C in benzene were measured by liquid scintillation counting using a LS spectrometer Quantulus 1220™, Teflon vials of 7.0 or 3.0 mL, and butyl-PBD (5 g/L) as a scintillator. We preset counting time, which allows us to obtain statistical uncertainty about 1%. Because we did not measure delta ^{13}C for our pine wood samples, we assumed delta ^{13}C to be –25.0‰ (average for wood) and used this value to calculate the radiocarbon concentration of all samples.

RESULTS AND DISCUSSION

All the obtained experimental data are given in Table 1. The ^{14}C data obtained for Rivne NPP together with the background data are shown on Figure 2. As a background, we use the ^{14}C in air data of Hua et al. (2013) which we continue with our experimental data for the time interval 2009–2020. These two data series intersect at the interval 2000–2009, from which we argue that they correspond to each other. At the same time, in the figures, we present all the data of both series to reflect their correspondence. ^{14}C in the annual growth of wood obtained for the vicinity of the Rivne NPP for the entire studied time interval, do not differ from the background within their uncertainty of ±1.0 pMC.

Experimental data obtained for the site located 3.5 km west-northwest of Chornobyl NPP and background data (described above) are shown in Figure 3. It can be seen that the data during the NPP operation period regularly exceed the background levels by 3.0–13.0 pMC when data for time interval 2001–2020 is indistinguishable from the background i.e., there are no further ^{14}C emissions because of the shut down of the Chornobyl reactors. The maximum level of ^{14}C excess agrees well with maximal levels reported for annual tree ring sampled for site 3.2 km east from Chornobyl NPP 15.9 pMC (1987) and 16.5 pMC (1990) (Buzynnyi and Talerko 2000). ^{14}C data for tree rings of pine tree for Kopachi site located 3.5 km southeast-south direction to Chornobyl NPP (Skripkin et al. 2005) are presented in Figure 3 as well for comparison. The general course of our Chornobyl NPP data presented here resembles the change over time of the data (Skripkin et al. 2005), but in some years there is a significant deviation and in different directions. Data are lower for 1988, 1992 and higher for 1994, 1995, which can be explained by the corresponding fluctuations in the prevailing wind direction for these years.

Table 1 Radiocarbon concentration (±1.0 pMC) in pine annual tree rings for background site (60 km, W to Kyiv), Rivne NPP (1.5 km, S-W) and Chornobyl NPP (3.5 km, N-W-W) locations.

Year	Background	Rivne	Chornobyl	Year	Background	Rivne	Chornobyl
1984			126.9 ± 1.1	2003	107.6 ± 1.0	107.5 ± 1.0	107.1 ± 1.0
1985			125.3 ± 1.1	2004	106.7 ± 1.0	107.0 ± 1.0	107.2 ± 1.0
1986		120.7 ± 1.1	182.7 ± 1.1	2005	106.3 ± 1.0	106.1 ± 1.0	107.0 ± 1.0
1987		118.3 ± 1.1	125.5 ± 1.1	2006	105.8 ± 1.0	105.8 ± 1.0	106.5 ± 1.0
1988		116.8 ± 1.1	121.0 ± 1.1	2007	105.1 ± 1.0	104.9 ± 1.0	106.1 ± 1.0
1989		117.7 ± 1.1	119.0 ± 1.1	2008	105.2 ± 1.0	105.6 ± 1.0	105.4 ± 1.0
1990		116.1 ± 1.1	120.0 ± 1.1	2009	104.8 ± 1.0	105.3 ± 1.0	104.5 ± 1.0
1991		115.3 ± 1.2	119.1 ± 1.1	2010	104.3 ± 1.0	104.0 ± 1.0	103.8 ± 1.0
1992		112.9 ± 1.1	115.9 ± 1.1	2011	104.0 ± 1.0	103.6 ± 1.0	103.8 ± 1.0
1993		113.5 ± 1.1	120.6 ± 1.1	2012	103.5 ± 1.0	103.3 ± 1.0	104.0 ± 1.0
1994		112.0 ± 1.1	124.5 ± 1.1	2013	103.2 ± 1.0	103.1 ± 1.0	103.7 ± 1.0
1995		111.5 ± 1.1	124.5 ± 1.1	2014	103.0 ± 1.0	103.0 ± 1.0	103.2 ± 1.0
1996		110.5 ± 1.1	117.5 ± 1.1	2015	102.5 ± 1.0	103.0 ± 1.0	102.6 ± 1.0
1997		110.3 ± 1.1	117.7 ± 1.1	2016	102.1 ± 1.0	103.2 ± 1.0	102.3 ± 1.0
1998		109.7 ± 1.1	114.2 ± 1.1	2017	102.1 ± 1.0	102.4 ± 1.0	101.9 ± 1.0
1999		109.3 ± 1.0	112.9 ± 1.0	2018	102.1 ± 1.0	102.7 ± 1.0	102.0 ± 1.0
2000	108.6 ± 1.0	108.9 ± 1.0	110.1 ± 1.0	2019	101.7 ± 1.0	102.1 ± 1.0	102.0 ± 1.0
2001	108.7 ± 1.0	108.3 ± 1.0	110.0 ± 1.0	2020	101.3 ± 1.0		101.7 ± 1.0
2002	107.8 ± 1.0	108.0 ± 1.0	110.8 ± 1.0	2021			101.1 ± 1.0

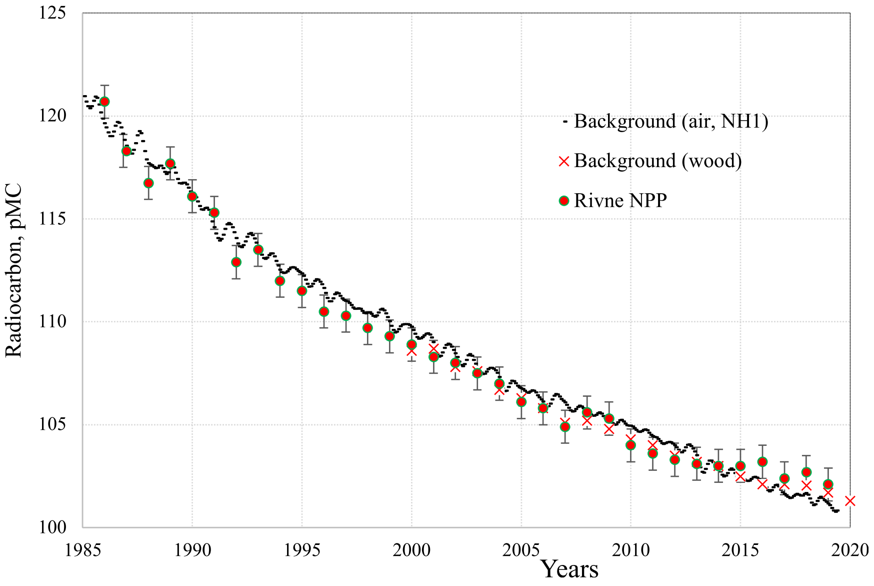

Figure 2 ^{14}C concentration in tree rings around Rivne NPP, background tree, and background (Hua et al. 2013).

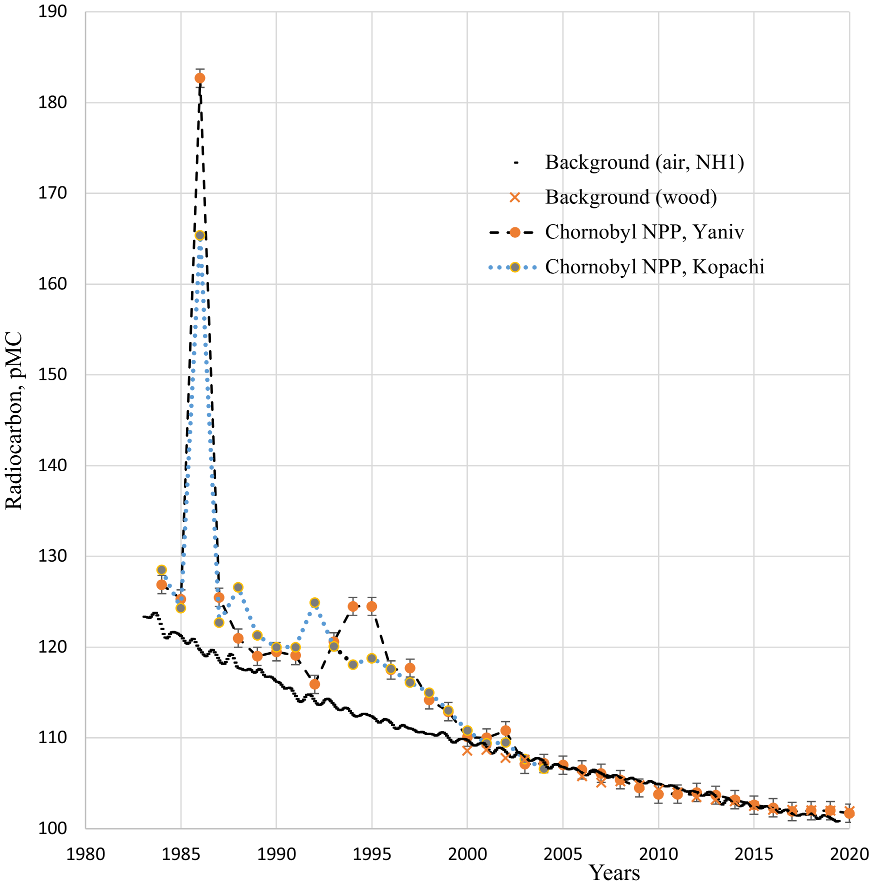

Figure 3 ^{14}C concentration in tree rings around Chornobyl NPP data (Skripkin et al 2005), background tree, and background (Hua et al. 2013).

It is known that the concentration in air of $^{14}CO_2$ released from the source decreases with distance from it. When considering the above-mentioned ^{14}C spatial distribution model (McCartney and Scott 1988) then the expected ^{14}C excess for the closer distance from the source (1.5 km vs. 3.5 km) is about 2.0–3.0 times higher, respectively. Accordingly, the $^{14}CO_2$ emissions for Rivne NPP are still significantly lower than the corresponding Chornobyl NPP emissions, especially considering that in the case of Rivne NPP the sampling location is much closer to the source (1.5 km vs. 3.5 km in the case of Chornobyl NPP).

CONCLUSIONS

^{14}C levels in the annual tree rings of pine at a distance of 1.5 km southwest of Rivne NPP do not differ from background levels ($^{14}CO_2$), which confirms that ^{14}C emissions from this NPP are insignificant. ^{14}C levels in the annual tree rings of pine at a distance of 3.5 km west-northwest of the Chornobyl NPP exceed the background levels by 3–13 pMC ($^{14}CO_2$) during the time of NPP operation, while after shutdown ^{14}C data for time interval 2001–2020 is indistinguishable from the background. The above-mentioned maximum level of ^{14}C excess agrees well with the maximal reported for 1987 and 1990 annual tree rings for the site 3.2 km east from Chornobyl NPP 15.9 and 16.5 pMC (Buzynnyi and Talerko 2000). The general course of our new Chornobyl NPP data presented here resembles the change over time of the data (Skripkin et al. 2005), but in some years there is a significant deviation of our data, which can be explained by the corresponding fluctuations in the prevailing wind direction for these years. $^{14}CO_2$ emissions of the Rivne NPP are significantly lower than the corresponding emissions of the Chornobyl NPP, especially considering that in this case the location of the investigated site is much closer to the source than in the case of Chornobyl NPP (1.5 km vs. 3.5 km), and expected ^{14}C excess should be 2.0–3.0 times higher.

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