ECOLOGICAL CHRONOLOGY OF NUCLEAR FUEL CYCLE SITES

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ABSTRACT. We compared ^{14}C levels in annual growth rings of pine trees around the Tomsk nuclear fuel reprocessing plant (NFRP) and the Chernobyl nuclear power plant (NPP). At the Chernobyl site, samples were taken from the control area (within a 30-km radius zone around the site) to a distance of 80 km. In Tomsk, we collected samples between 8–10 km and 10–15 km from the site, taking into account prevailing wind directions. Background samples were collected 200 km from the plant. Samples were converted to benzene and counted in an ultra-low-level LSC Quantulus 1220™. Because of the Chernobyl accident, a signal can be detected in the background of routine plant operation. Comparison with the Tomsk data suggests that the routine discharges from Tomsk are more significant than the discharge from the Chernobyl accident. We estimated Tomsk NFRP annual discharge level at up to 30–45 TBq ^{14}C from 1985 to 1988.

INTRODUCTION

According to data published by the United Nations (1993), total ^{14}C emissions from the nuclear industry was equivalent to 150 TBq in 1980, 360 TBq in 1985, 580 TBq in 1990, and by the year 2000, will reach almost 1000 TBq (Otlet, Fulker and Walker 1992). Some controversy exists on the contribution of different types of nuclear facilities to total ^{14}C discharges. According to Bush, White and Smith (1983), nearly 70% of the total ^{14}C discharges in the year 2000 will be from fuel reprocessing. Studies conducted by McCartney and Scott (1988a,b) around the Sellafield Nuclear Fuel Reprocessing Plant (NFRP) give figures up to 20 TBq for the annual discharges from the site. We provide details of an analogous study (Kovalykh *et al.* 1994) around Tomsk, a fuel reprocessing plant situated in western Siberia (57° N), which has been operating since 1953, using uranium-graphite reactors (ADE-4.5). Tree rings were sampled from 8–10 to 10–15 km southeast and northwest of the plant. We also obtained 1994 data for grass to the southeast. At the same time, a similar study was conducted around the Chernobyl power plant, in which RBMK-1000 reactors were used. The first data relating to ^{14}C releases from Chernobyl were reported by Salonen (1987), although earlier data on tree rings were also published (Buzinny *et al.* 1992, 1993). We sampled pine trees in 1989 and in 1992–1994 within the control area (boundary 30 km) and out to 100 km. Background samples were collected over 200 km from the site. Annual tree rings were sampled from 1979–1993. For the 1986 samples, the rings were split into early and late wood to sensitize detection of any effect from the Chernobyl accident.

METHODS

The tree-ring samples were given preliminary chemical preparation before counting in proportional (Gliwice) and liquid scintillation (LS) counters (Kiev). Samples for LS were counted in an ultra-low-level Quantulus 1220™ counter using PPO and POPOP as scintillators and aluminium Teflon® and copper Teflon® vials. Results were corrected for both fractionation and quenching effects. Oxalic acid was used as the primary standard. Samples of 1986 rings were separated into early and late

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wood in order to maximize the sensitivity of the method to detect any effect of the Chernobyl accident. More precise steps were also used for some tree-ring samples when both early and late wood were split into two equal parts for the same reason.

RESULTS

Chernobyl Data

Figure 1 shows results from the study of long-term variations in one tree, as well as the 1986 result for three other trees located at distance of 2.4 and 4 km from the site. The long-term trend from 1965–1978 is very similar to the global ^{14}C levels in the atmosphere. The data from 1979–1985 and 1987–1989 show elevated levels corresponding to the operational activity of the plant. The results for 1986 give specific activities in the range from 720 to 6533 Bq kg^{-1}, and show a high degree of local spatial variation.

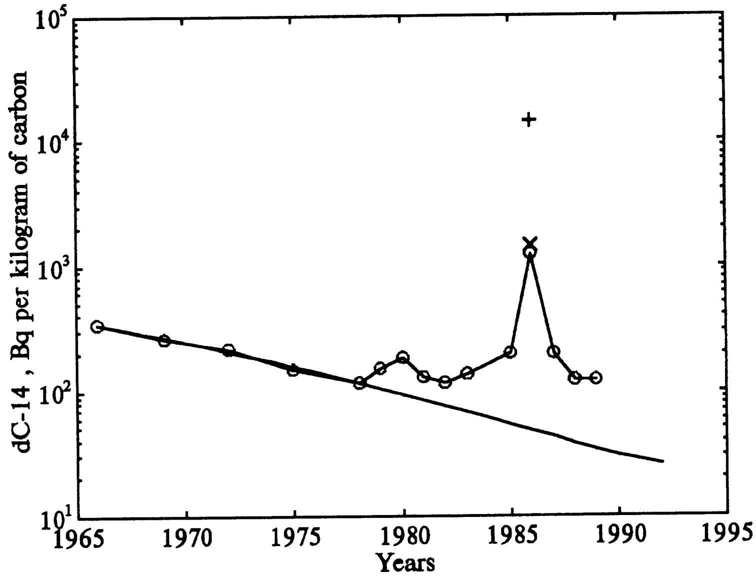

Fig. 1. ^{14}C specific activity in pine tree rings collected near Chernobyl NPP, Bq kg^{-1} C (Buzinny *et al.* 1992). + = "Red Forest", 4 km W-N; X = "Red Forest", 2.5 km W; O = "Red Forest", 6 km W-S; — = extrapolation of years 1966–1978.

For subsequent sampling around Chernobyl, we used an aerial survey of gamma radiation. The background sample was obtained 200 km south of Chernobyl. Table 1 shows the 1986 results for trees around Chernobyl as well as for background samples. There is significant variation in ^{14}C level over the samples; thus, we developed an optimized sample preparation method in which the early and late wood are separated for some of the trees. The samples were then split in half. We believe this procedure may increase the detection of excess ^{14}C (Table 2).

We determined the maximum ^{14}C level in samples collected 4 km northwest of Chernobyl, corresponding to the specific activity of 6500 Bq kg^{-1} C. We estimated the average ^{14}C concentration in air during the release to be 50 Bq m^3 (averaged over a 14-day discharge period). This figure is subject to variation due to changes in wind direction and weather conditions; thus, it is not unreasonable to hypothesize maximum levels to be 100–1000 times higher than the mean level. Table 1 clearly shows considerable local variation in ^{14}C levels, and that increased sensitivity has been obtained through our sampling method. It is reasonable to assume other discharges may behave like CO_2. Therefore, the use of this method for reconstructing past discharges seems possible, something we are currently examining.

TABLE 1. ^{14}C concentration in 1986 annual tree-ring samples collected around Chernobyl NPP. Uncertainty is *ca.* 3.5 Bq kg^{-1} C for early and late wood samples, *ca.* 3% for other samples.

No.	Site	Direction	Distance (km)	1986	1986 (E)*	1986 (L)†
1	"Red Forest"	NW	4.0	857.0	–	–
2	Tree nursery	W	2.5	6533.0	–	–
3	Leljev	SW	6.0	720.0	–	–
4	Paryshev	SE	10.0	327.0	350.7	303.3
5	Sachan	N	12.0	308.6	345.5	271.2
6	Janov	W	2.0	9700.0 (E)	17,130.0 (1)‡	302.5 (2)‡
7	Janov	W	2.0	302.1 (L)	305.0 (1)	300.2 (2)
8	Tarastcha§	S	200.0	272.6 (E)	274.3 (1)	270.8 (2)
9	Tarastcha	S	200.0	268.7 (L)	268.2 (1)	269.2 (2)

*Early wood
†Late wood
‡1 or 2 indicates early and late wood were divided into two equal parts in the order L1, L2, E1, E2
§Tarastcha=background sample

TABLE 2. Excessive ^{14}C (Bq kg^{-1} C above natural level) in annual rings of trees around nuclear reprocessing plant Tomsk-7, Russia Tomsk samples = 10 km NE; Tomsk 2 samples = 10–15 km SW.

Year	Tomsk	Year	Tomsk	Tomsk 2	Year	Tomsk	Tomsk 2	Year	Tomsk	Tomsk 2
1949	–6.6	1961	113.0	--	1973	143.7	101.7	1984	155.5	64.9
1950	2.7	1962	128.3	--	1974	147.6	101.2	1985	173.2	57.7
1951	–3.9	1963	198.6	--	1975	154.8	89.0	1986	167.5	62.7
1952	2.3	1964	239.3	--	1976	159.1	109.6	1987	160.9	56.8
1953	5.4	1965	245.4	--	1977	149.8	90.8	1988	171.8	56.5
1954	14.8	1966	221.3	--	1978	153.7	81.9	1989	163.2	--
1955	13.4	1967	200.0	--	1978	153.7	81.9	1990	160.7	--
1956	16.6	1968	170.7	--	1979	168.7	67.6	1991	157.1	--
1957	37.5	1969	146.9	129.4	1980	177.1	75.6	1992	153.0	--
1958	49.0	1970	153.0	126.2	1981	164.6	67.0	1993	115.3	--
1959	76.5	1971	143.5	121.0	1982	157.3	75.6			
1960	92.2	1972	155.9	113.5	1983	145.1	55.6			

Tomsk-7 Data

We determined the variability in ^{14}C-specific activity in tree rings and in the atmosphere at the Tomsk site from 1953–1993. The Tomsk results can be compared to other atmospheric data (Kaimei, Youneng and Fan 1992) for a background site. Both data series are shown in Figure 2. At Tomsk-7, the ^{14}C discharges have been increasing since 1970 and have resulted in a ^{14}C level exceeding 120 Bq kg^{-1} C in the tree rings 10 km from the site. The 1993 data show evidence of a decrease in ^{14}C discharges.

We have also employed the data to determine the discharge levels retrospectively, using the data of McCartney and Scott (1988a,b) to determine a coefficient relating excess ^{14}C in the tree rings to discharge. Our justification for this is that the spatial distribution of the site is similar to Sellafield (*e.g.*, in 1985, 20–30 Bq kg^{-1} C, corresponding to a discharge of 7 TBq). Using this approach, we estimate the coefficient to be 0.23–0.35 TBq a^{-1} Bq^{-1} kg. In the future, more experimental work will be carried out to define exact spatial distribution for the site. Using this coefficient, we estimate the highest Tomsk discharge at 1.5–2.5 times higher (Fig. 3) than the relative ^{14}C maximum at Sellafield in 1981. We estimate maximum annual discharges to be 31 TBq a^{-1} in 1988 and the total discharge during the operational period to be 450–620 TBq.

1994 grass samples collected at 5–6 km southwest (wind direction) yield an excessive ^{14}C level of 54.9 ± 4.6 (Bq kg^{-1} C above natural level). This establishes a decrease in the level of ^{14}C discharges since 1993 (compare the 1988 value of 56.5 Bq kg^{-1} at 15 km southeast of T2).

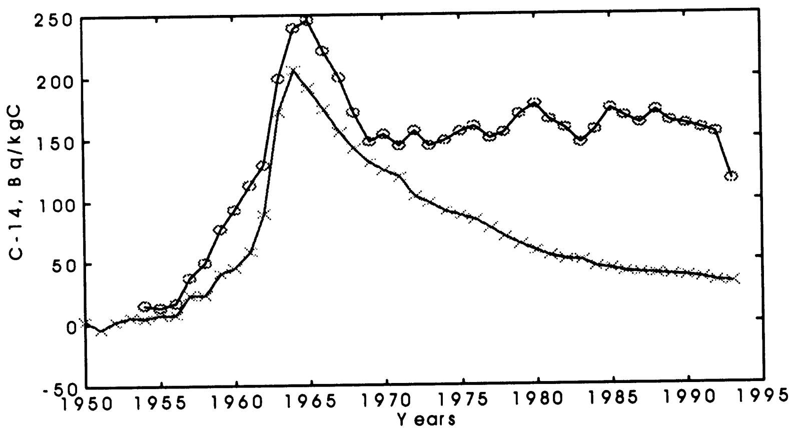

Fig. 2. ^{14}C concentration (o) of annual pine tree rings (Bq kg^{-1} C) around the Tomsk-7 nuclear reprocessing plant (Russia), relative to background. Pre-1950 AO background taken as 0. The bomb ^{14}C curve (x) is also shown from a background site (Kaimei *et al.* 1992).

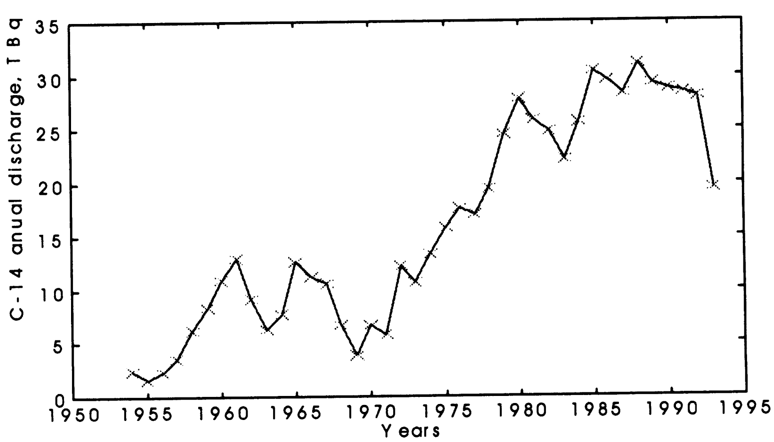

Fig. 3. Reconstructed annual ^{14}C discharges for Tomsk (details in text)

CONCLUSION

We conclude that, using the methodology described above, we can determine excess ^{14}C levels to at least 30 km from the Chernobyl site, and can extend our study area to other sites on a wider scale. At both Tomsk and Chernobyl, effects of the ^{14}C discharges are local and spatially varied. The comparison of Tomsk and Chernobyl shows that routine releases at Tomsk are more significant than those at Chernobyl. However, as a result of the accident at Chernobyl, and taking into account wind direction and weather variation, the maximum ^{14}C atmospheric concentration was 100–1000 times higher than the average level of 50 Bq m^{-3}. These high levels are found in the early wood of 1986 tree rings. Because the behavior of other gases is similar to that of CO_2 discharged during the accident, we should also be able to reconstruct and make a radiological assessment of the discharge levels of those gases. We are planning further studies to provide more detail about the distribution of excess ^{14}C around Chernobyl and to establish discharge records for both sites.

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