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TREE RING RECORD OF RADIOCARBON EMISSION FROM NUCLEAR FUEL REPROCESSING PLANT TOMSK–7

Summary. Excessive radiocarbon concentrations in the annual tree rings due to operation of nuclear fuel reprocessing plant Tomsk–7, situated in west Siberia (57°N, 85°E), are presented in that report. The plant was put in operation in 1953. The technology there uses uranium-graphite reactors ADE-4.5. Two pine trees were used to reconstruct radiocarbon levels around the site: at the distance of 10 km SE and at the distance of ca 15 km NW. The T1 tree covers the period from 1949 to 1992, while T2 covers the period from 1970 to 1989. Excessive ^{14}C data received are up to 130 Bq per kilogram of carbon in 1985–88 for distance of 10 km from the site, which corresponds to up to 30–45 TBq ^{14}C discharge per year relatively.

ZAPIS EMISJI RADIOWĘGLA Z ZAKŁADÓW PRZERÓBCZYCH PALIWA JĄDROWEGO TOMSK-7 W SŁOJACH ROCZNYCH PRZYROSTÓW DRZEW

Streszczenie. Artykuł przedstawia wyniki pomiarów koncentracji izotopu ^{14}C w słojach rocznych przyrostów drzew rosnących w pobliżu zakładów ptrzeróbczych paliwa jądrowego Tomsk-7, zlokalizowanych w pobliżu miasta Tomska na zachodniej Syberii (57°N, 85°E). Zakłady zostały uruchomione w 1953 roku; wyposażone są w reaktory uranowo- grafitowe ADE-4.5. Do badań użyto dwóch pni sosny: pień drzewa T1 ściętego w odległości 10 km na SE od zakładów, obejmujący lata 1949–1992, oraz pień drzewa T2, ściętego w odległości 15 km na NW od zakładów, obejmujący zapis z lat 1970–1989. Stwierdzono nadwyżkę izotopu ^{14}C na poziomie 130 Bq/kg w latach 1985–1988 w odległości 10 km od zakładów, co odpowiada rocznej emisji izotopu ^{14}C na 30-45 TBq.

1. Introduction

Main sources of artificial radiocarbon in atmosphere and biosphere are nuclear power plants, nuclear fuel reprocessing facilities and nuclear reactors in research institutions. Environmental radiocarbon levels around various types of nuclear installations have been monitored in numerous sites in Europe using atmospheric and plant samples.

Carbon isotopes accumulated in annual tree rings in the vicinity of nuclear facilities reflect changes of isotope concentrations in the atmosphere and may serve as precise tool for monitoring and retrospective evaluation of radiocarbon emission during normal operation of nuclear installations and emission associated with more or less severe failures or accidents. According to data from UN Economic Commission for Europe (1983), quoted by Otlet et al. (1992), total radiocarbon emission from nuclear industry was equivalent to 150 TBq in 1980, 360 TBq in 1985, 580 TBq in 1990, and in 2000 it will reach almost 1000 TBq. There is a distinct controversy about contribution of different types of nuclear facilities to the total radiocarbon discharges. According to Bush et al. (1984) nearly 70% of the total radiocarbon discharge in year 2000 will be from fuel reprocessing (Otlet et al., 1992).

Another estimates, presented by UNSCEAR (1988) suggest that the releases from the energetic nuclear reactors are considerably greater. Detailed studies of McCartney et al. (1988) of radiocarbon emission from nuclear fuel reprocessing facility in Sellafield indicate average radiocarbon emission at level 7.7 TBq per year in 1970–79 and at level 12.3 TBq per year in 1980–85, with maximum values reaching 20 TBq per year in 1983 and 1984 (McCartney et al., 1988a,b).

Comparison of these figures with estimates of global radiocarbon discharges indicates for important contribution of fuel reprocessing facilities to the global radiocarbon inventory. It seems obvious that any evaluations of the long–term impact of radiocarbon emission from nuclear industry on the public health in both local and global scales need more experimental data obtained from detailed studies of selected specific sites.

In this report we present results of studies of radiocarbon concentration in the vicinities of nuclear fuel reprocessing plant Tomsk-7 situated in west Siberia (57°N, 85°E). The plant was put in operation in 1953. The technology there uses uranium-graphite reactors ADE-4 5.

Two trees of pine were used to reconstruct radiocarbon levels around the site: T1 was cut at the distance ca 10 km NE (prevailing winds direction), and T2 at the distance of ca 15 km SW. The T1 tree covers period from 1949 to 1992, while T2 covers period from 1970 to 1989. Three samples of 1991 grass were also examined.

2. Methods

Annual tree ring samples were separated from collected trees at the Tree Ring Laboratory of the Tomsk State University and Gliwice. Individual tree ring samples were subjected to preliminary chemical treatment consisting or extraction of volatile compounds in a Soxhlet extractor. Then samples were converted either to carbon dioxide (Gliwice) or benzene (Kiev) for radiocarbon determinations. Measurements of radiocarbon activity were performed using liquid scintillation counting (Kiev) or carbon dioxide filled proportional counters (Gliwice) NBS Oxalic Acid SRM 4990 was used as a reference sample of modern radiocarbon activity. All results of radiocarbon activity measurements were corrected for isotopic fractionation using experimentally determined values of $\delta^{13}C$ in analysed sample as well as quench correction for liquid scintillation counting.

3. Results

Figure 1 shows the simplified map of the study area with indicated sampling points. According to prevailing wind direction of S-W to N-E we studied ^{14}C level in two trees situated on the distance of 8-10 km (N-E) and 10-15 km (S-W) to Tomsk-7 NFRP. Data received for ^{14}C specific activity in annual rings for the period of 1953 to 1993 were already discussed together with background values (McCartney et al., 1988a,b; Dai Kaimei et al.).

The levels of excessive ^{14}C (above natural level) are shown on Figure 2. Results of measurements of radiocarbon concentration in all analysed samples are shown on Figure 2 and Table 1 in form of $\Delta^{14}C$ (Bq per kilogram) values that denote relative excess of ^{14}C above the normal level in uncontaminated biosphere.

Since 1970 the levels of ^{14}C discharges increased for Tomsk–7 and led to ^{14}C exceeding 130 Bq per kilogram of carbon in annual rings on the distances of 10 km from the site. Figure 3 shows same data received after substraction of background levels for both of trees.

Lover line on Figure 1 shows atmospheric values over Asia derived from tree ring studies of Dai Kaimei et al. (1992) and may be used as reference level in analysis of the magnitude of local effects associated with radiocarbon emission from Tomsk–7 nuclear installations. The record radiocarbon variations obtained in T1 locality clearly reveals three different periods: during first four years (1949-52) radiocarbon concentrations in T1 annual rings are very close to uncontaminated biosphere before the nuclear period and then, since mid-fifties, they are systematically higher than corresponding regional level recorded by Dai Kaimei et al. (1992). In the third period since 1970 till the present, local Δ ^{14}C values remain at approximately constant level of ca 150 Bq per kilogram with

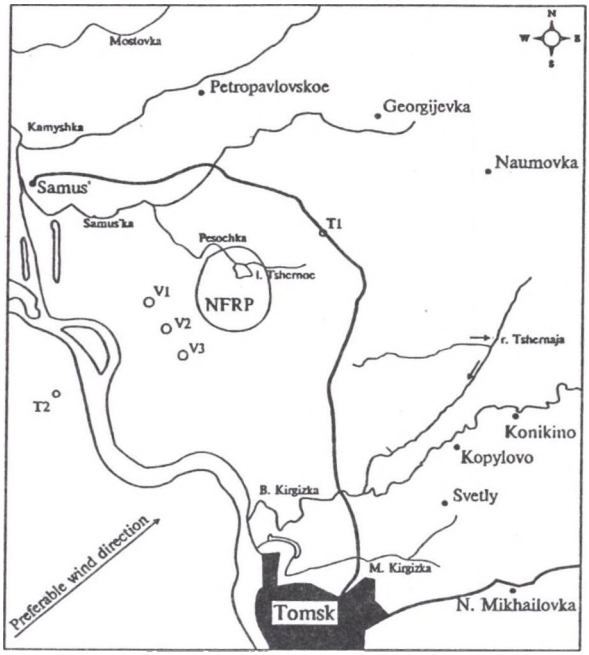

Fig. 1. Tomsk NFRP sampling map

Rys. 1. Mapa okolic Tomska z miejscami poboru próbek

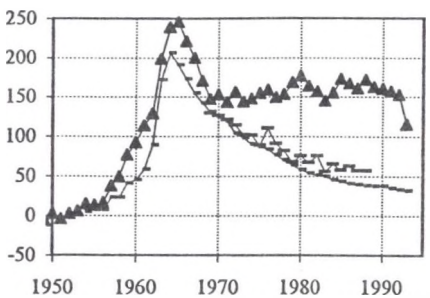

Fig. 2. Excessive ^{14}C (Bq per kilogram of carbon above natural level) in annual rings of trees around the nuclear reprocessing plant Tomsk-7, Russia. From above-down: T1 - N–E distance 10 km, T2 - S–W, 15 km, backgroun level

Rys. 2. Nadmiar ^{14}C (w Bq/kg węgla ponad poziam naturalny) w słojach rocznych przyrostów drzew rosnących wokół zakładów przetwórczych paliwa jądrowego Tomsk-7 w Rosji. Górna krzywa T1 kierunek N-E w odległości 10 km, dolna krzywa T2 kierunek S-W, w odległości 15 km i wartość tła

Table 1

Excessive ^{14}C (Bq per kilogram of carbon above natural level) in annual rings of trees around the nuclear reprocessing plant Tomsk-7, Russia. Samples: Tomsk- N-E distance 10 km, Tomsk 2 - S-W,10-15km

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			

some minor short–tern fluctuations. The corresponding record obtained in T2 vicinity is limited to the period of the last 20 years (1970–90) and show decreasing global trend of radiocarbon levels with distinct contribution of locally produced radiocarbon. On average the local excess of radiocarbon in locality T2 during the study period may be estimated as equivalent to $\Delta^{14}C$ increase by ca 25 Bq per kilogram.

1994 grass samples are collected aproximatelly at distance 5–6 km S–W direction across wind direction. Data obtained show excessive ^{14}C level of 54.9±4.6 (Bq per kilogram of carbon above natural level). That establish decrease of ^{14}C discharges level since 1993 (compare 56.5 Bq per kilogram for T2 in 15 km S–W for 1988).

4. Conclusion

Significant levels of excessive ^{14}C variation are observed for vicinity of Tomsk Nuclear Fuel Reprocessing Plant (prevailing wind direction) during operation period (up to 130 Bq/kg for $\Delta^{14}C$). It corresponds to serious level of $^{14}CO_2$ release attained of ca 30-45 TBq per year in maximum.

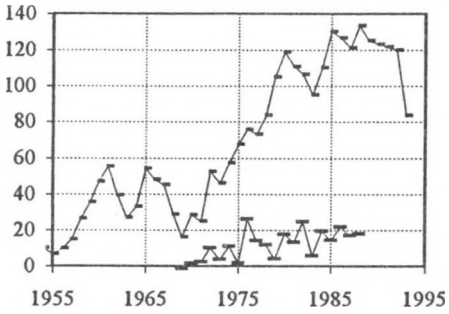

Fig. 3. Excesive ^{14}C (Bq per kilogramof carbon above bacground level) in annual rings of trees around the nuclear reprocessing plant Tomsk–7, Russia. From above–down: T1–N–E distance 10 km, T2–S–W distance 15 km

Rys. 3. Nadmiar ^{14}C (w Bq/kg węgla ponad poziam naturalny) w słojach rocznych przyrostów drzew rosnących wokół zakładów przetwórczych paliwa jądrowego Tomsk–7 w Rosji. Górna krzywa T1 kierunek N–E w odległości 10 km, dolna krzywa T2 kierunek S–W, w odległości 15 km

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Streszczenie

Poziom koncentracji radiowęgla wokół różnego typu instalacji jądrowych został przebadany w licznych miejscach w Europie. Wykorzystano przy tym atmosferyczny dwutlenek węgla oraz próbki roślin. Izotopy węgla zawarte w pierścieniach drzew rosnących w otoczeniu instalacji jądrowych są odbiciem zmian koncentracji tychże izotopów w atmosferze i mogą służyć jako precyzyjne narzędzie kontroli i oszacowań emisji radiowęgla podczas normalnej pracy instalacji jądrowych oraz emisji towarzyszącej sytuacjom incydentalnym.

Praca zawiera wyniki badań koncentracji radiowęgla w otoczeniu elektrowni jądrowej Tomsk–7, znajdującej sięw w zachodniej Syberii (57°N, 85°E). Elektrownia rozpoczęła pracę w 1953 roku na bazie reaktora uranowo–grafitowego ADE–4.5. Do rekonstrukcji koncentracji radiowęgla w otoczeniu elektrowni wykorzystano dwa pnie sosny: pień T1, pochodzący z drzewa ściętego w odległości około 10 km, w kierunku NE (przeważajacy kierunek wiatrów), oraz pień T2, pochodzący z drzewa ściętego w odległości około 15 km na SW. Pierścienie pnia T1 obejmują okres czasu od 1949 do 1992 roku, zaś pnia T2 – od 1970 do 1989 roku. Ponadto przebadano trzy próbki traw z 1994 roku.

Roczne pierścienie drzew zostały wyseparowne z pnie w „Tree Ring Laboratory" Uniwersytetu w Tomsku. Pojedyncze próbki podlegały traktowaniu chemicznemu w aparacie Soxhleta w celu usunięcia lotnych związków organicznych, a następnie przetworzone zostały na dwutlenek węgla (Gliwice) lub benzen (Kijów) w celu dokonania pomiarow koncentracji radiowęgla. Pomiary zostały wykonane metodą scyntylacyjną (Kijów) oraz za pomocą liczników proporcjonalnych (Gliwice). Wyniki wykazały, że począwszy od połowy lat sześćdziesiątych koncentracja radiowęgla w pierścieniach rocznych rdzenia T1 narastała osiągając wartość 130 Bq na kilogram węgla, co odpowiada produkcji radiowęgla w ilości 30–45 TBq na rok.