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^{14}C TRACED IN KRAKÓW AFTER THE CHERNOBYL ACCIDENT

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ABSTRACT. Results of the ^{14}C measurements in atmospheric CO_2 in the first half of 1986 are presented. CO_2 samples were systematically collected in Kraków in two-week cycles and, after conversion to benzene, measured in a liquid scintillation spectrometer. ^{14}C activity and $^{13}C/^{12}C$ ratio are reported as $\delta^{14}C$ and $\delta^{13}C_{PDB}$, respectively. For about three weeks after April 26, 1986 (the Chernobyl accident) an increase of ~9% above the normal level for Kraków was observed. A rough estimate of the ^{14}C release to the lower atmosphere during the accident gave a value 900 Ci, which is $\sim 1.8 \times 10^{-5}$ of the total activity released to the atmosphere.

INTRODUCTION

We have been measuring carbon isotope concentration in the Kraków region for about three years. Results of the first two years are presented in Kuc (1986). The sampling location is in Kraków (50° 3′ N, 19° 54′ E) not far from the city center and close to recreation and sports grounds. Kraków and its suburbs belong to a densely populated and industrialized area.

An accident in the Chernobyl Nuclear Power Station made monitoring of natural radioisotope background in Spring 1986 especially interesting. During this period, besides ^{14}C in atmospheric CO_2, other radioisotopes were also monitored (Duliński *et al,* in press).

SAMPLING TECHNIQUE

For the last three years samples were continuously collected at two-week intervals at ca 20m above the ground level, on the roof of our Institute building, where an inlet to the sampling assembly is mounted. Collection is performed by continuously passing the pumped air through a column with silicagel (ca 800g) for trapping water and then through a stainless steel container filled with a molecular sieve, 4 Å, in which CO_2 is separated and sorbed. The volume of the pumped air, usually ca $15m^3$ per sample, is measured by a gas meter at the inlet. The flow rate is controlled by a flow indicator mounted at the outlet. A simple water pump is used for pumping. In case of a drop in tap-water pressure, pumping is switched to an electrically driven membrane pump.

Recovery, cleaning, conversion to benzene, and finally measurement of CO_2 are after the procedure routinely used (Grabczak *et al,*1983).

^{14}C activity is reported as $\delta^{14}C$ according to the notation used in radiocarbon laboratories, and necessary $\delta^{13}C$ mass spectrometer measurements are carried out. Details of technical problems are described in Kuc (1986).

^{14}C FROM ATMOSPHERIC CO_2

Measurement results of ^{14}C in the form of $\delta^{14}C$ together with $\delta^{13}C_{PDB}$ are listed in Table 1, and a plot of the $\delta^{14}C$ *vs* time is shown in Figure 1.

An abrupt "jump" in $\delta^{14}C$ values was noticed in two samples, POW-88 and POW-89, collected within four weeks from April 21 to May 19. These two samples show a distinctly higher level of atmospheric ^{14}C activity from neighboring ones.

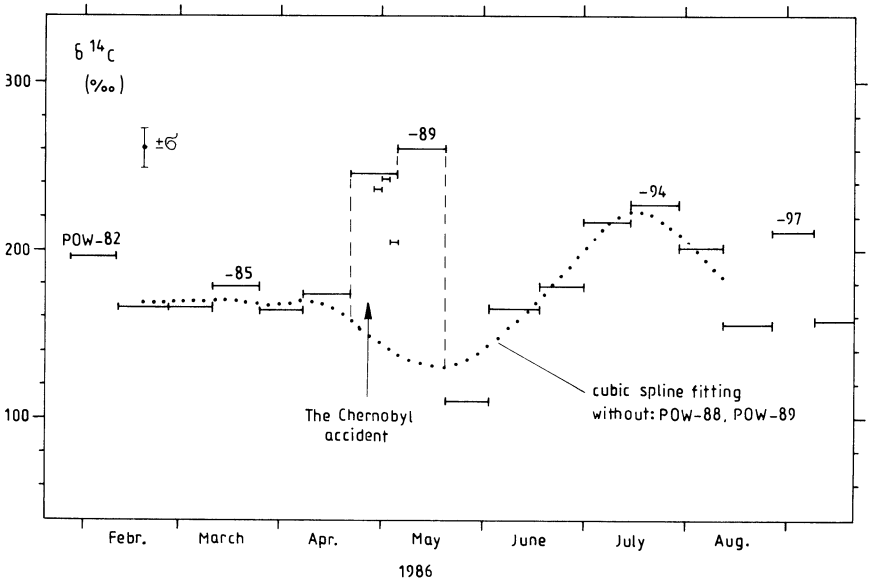

Fig 1. Time plot of $\delta^{14}C$ of atmospheric CO_2 collected in Kraków, February–September 1986

For the seven-day period, April 28 to May 5, three additional samplings of ^{14}C were taken using the second sampling assembly. The duration of each collection was about two days. The basic aim of these additional measurements was to obtain control results to those obtained in routine monitoring and to detect, if possible, the highest ^{14}C activity. Results are marked in Figure 1 as three short bars.

TABLE 1
Isotopic composition of carbon in atmospheric CO_2 in Kraków after the Chernobyl accident

Sample lab no	Colln date (year, month, day)	$\delta^{14}C$ (‰)	$\delta^{13}C$ (‰)
POW-82	1986 1/27–2/10	197 ± 10	−10.16
-83	2/10–2/24	166 ± 11	−9.87
-84	2/24–3/10	166 ± 11	−10.66
-85	3/11–3/24	179 ± 9	−9.95
-86	3/24–4/7	163 ± 9	−9.68
-87	4/7–4/21	174 ± 9	−9.76
-88	4/21–5/5	247 ± 9	−9.99
-89	5/5–5/19	261 ± 11	−9.42
-90	5/21–6/2	120 ± 9	−9.72
-91	6/2–6/16	167 ± 11	−8.90
-92	6/16–6/30	179 ± 11	−8.64
-93	6/30–7/14	218 ± 12	−9.08
-94	7/14–7/28	228 ± 12	−9.04
-95	7/28–8/11	202 ± 11	−9.96
-96	8/11–8/26	156 ± 11	−9.32
-97	8/26–9/8	212 ± 12	−8.93
-98	9/8–9/22	158 ± 8	−9.45

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In general, slightly lower values of the two-day collections compared to the two-week sampling most likely come from the different sampling height above the ground level. In the case of the three collections, sampled air was sucked in at the height of ca 2m where the influence of CO_2 respired by green plants and that generated by decomposition of organic matter was much greater.

The third and last result is remarkably lower and correlates well with the plot of total β^- activity of air particulates which shows the local minimum between May 1 and 5 (Duliński *et al*, in press). The same tendency is visible for atmospheric moisture.

ESTIMATION OF THE ^{14}C RELEASE AT CHERNOBYL

It would be interesting to assess the ^{14}C activity released into the atmosphere during the Chernobyl accident, even with an accuracy that can be made to an order of magnitude only.

The laboratory closest to the accident site that could measure atmospheric $^{14}CO_2$ was located in Kraków, in a straight distance of ca 740km from Chernobyl.

Results presented in the preceding section enable us to separate the net effect detected in Kraków, assuming that only samples POW-88 and POW-89 (collected from April 21 to May 19) were contaminated by Chernobyl. All the rest represent the normal level of ^{14}C in the Kraków atmosphere and these values can be smoothed, eg, by spline functions (Reinsch, 1967, 1971). The smooth curve is taken for further calculations as the background level of ^{14}C.

After drawing a histogram (Fig 1) the calculated excess of the ^{14}C activity is ca 10% of modern carbon. This figure represents a mean value for a two-week sampling period (^{14}C was collected for two weeks at a constant speed) and is close to the summer–winter variation (Kuc, 1986).

The duration of the immediate Chernobyl contamination was not longer than 23 days (April 26 to May 19) and certainly not shorter than ca 9 days. The ^{14}C activity after May 5 is very close to that observed after April 21 which indicates the inflow of the similarly contaminated air masses also in the second week of May. The long lasting release of ^{14}C during the accident can be explained by production of $^{14}CO_2$ as a result of burning the reactor graphite which comprises ^{14}C produced in (n, γ) reaction with ^{13}C.

It is obvious that "Chernobyl" $^{14}CO_2$ above Polish territory is only a

TABLE 2
Some parameters of the radioactive release at Chernobyl

Duration of the radioactive emission	~10 days
Estimated total radioactivity released	50 MCi 3.5% of the present in the reactor (USSR report, 1986)
Estimated release of ^{14}C	900 Ci
Increase of the ^{14}C level in atmospheric CO_2 in Kraków	~10 pmc = ~9% of background
Duration of the ^{14}C contamination in Kraków	$\lesssim$3 weeks

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fraction of the total release. According to the Polish meteorological service, the radioactive cloud reached a height of ~3km and prevailing wind directions indicated that Poland received about one day of the "Chernobyl emission," while the duration of the radioactive releases was approximately ten days.

If we assume that 10% of the total Chernobyl releases caused an observed 9% increase of ^{14}C above the area close to the Polish territory (~300,000km^2) we can roughly estimate the total ^{14}C "production" during the accident. The value obtained in this way is 900 Ci, which compared with the activity of all other radioisotopes estimated in the USSR report (1986) to ~50 MCi, is negligibly small (1.8×10^{-5}). Some parameters of the radioactive release are comprised in Table 2.

It should be noted that in the above discussion only the "immediate ground level" contamination was considered by using the simplest model, disregarding the radioactivity penetrating the upper part of the atmosphere. The accuracy of the obtained value is expected to be within one order of magnitude.

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