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RECENT ^{14}C ACTIVITY IN THE ATMOSPHERE, "CLEAN AIR" AND THE CHERNOBYL EFFECT

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ABSTRACT. Although the Chernobyl accident caused increased ^{14}C levels in certain areas, it has been difficult to prove that it had any effect in two areas of Sweden and one on Svalbard (Spitsbergen), where the precipitation and wind conditions were such that the γ-active fall-out was negligible. Knowledge of the steady regional decrease and annual variations at high latitudes, where the pollution from fossil fuel is less than in central Europe, is essential for global studies of the CO_2 cycle. The present ^{14}C excess is a net effect of the ^{14}C supply, mainly from tests of nuclear weapons, and dilution, by ^{14}C-free, fossil-fuel consumption. In Sweden, at these northern latitudes, the ^{14}C excess is steadily slightly higher than for "clean air" in central Europe. Annual variations are also smaller in Sweden and Svalbard than in central Europe. The normal ^{14}C excess on Svalbard is slightly less than in Sweden. Detailed results, especially from autumn 1984 to autumn 1987, are given for atmospheric CO_2 collected in northern Sweden (Abisko) and on Svalbard (Kapp Linné) and for some atmospheric samples and plant material collected ca 50km east of Uppsala, very close to heavily polluted areas.

INTRODUCTION

Carbon dioxide from the atmosphere was collected at Abisko in northern Sweden and at Kapp Linné on Svalbard (Spitsbergen) for almost 30 years and measured at the conventional ^{14}C laboratory in Uppsala. During the last 10 years, samples, mainly plant material, such as tree leaves, were also collected at Smara, to the east of Uppsala. These records can be used in studies of both global ^{14}C variations and local and regional deviations from the global pattern.

COLLECTION AND MEASUREMENTS

Carbon dioxide is regularly collected by static absorption in diluted (0.5N) NaOH on trays fitted with thermal bimetal switches, adjusted to prevent freezing. Each tray is also equipped with a roof resting on perforated walls to prevent birds, insects and rain from reaching the NaOH while still allowing the CO_2 to be absorbed. The tray at Kapp Linné (78°04′N, 13°38′E) is on top of a small house at the radio and meteorological station. The tray at Abisko stands on top of a building at the Abisko Naturvetenskapliga Station (68°20.5′N, 18°49.3′E) 390m asl. The collection at Smara (59°53′N, 18°25 1/2′E) is made in the vicinity of a summer house. The NaOH on the trays is usually exposed to the atmosphere for three days and nights. The isotopic fractionation is naturally significant. The NaOH solution is reduced in the laboratory by a factor of ca 10 by boiling in a round, long-necked bottle with a narrow tube fitted to the end of the neck, yielding a 0.3m "chimney." Any exchange with the ambient atmosphere is thus prevented, and a suitable volume is achieved. The CO_2 is then liberated by adding HCl to the solution.

The ^{14}C activity is measured in a proportional counter filled with CO_2. Only one counter was used for the present study. The samples from the different collection sites were measured alternately, so that any difference between them cannot be explained by postulating a bias between the detectors. The observed ^{14}C trend from autumn 1984 to autumn 1987 is also real, since the samples from 1984 were measured immediately after those from 1987.

$\delta^{13}C$ is measured by normal mass spectrometry, and the ^{14}C content is normalized in the conventional manner. In this study, no consideration was given to the fact that part of the CO_2 was of fossil origin with a $\delta^{13}C$ value ca 18‰ below that of the CO_2 in the atmosphere.

The plant material from Smara mostly consisted of tree leaves and twigs from identical growing periods. The leaves and twigs were first harvested about two months after leafing, from either stubs or a tree felled for this purpose. All leaves and twigs grown during the collection year were then removed. The next harvest from the stubs took place in the autumn. In 1986, rhubarb was also collected, since these plant leaves were still smaller than normal eggs when the accident occurred but grew to ca $5dm^2$ by harvest time on May 4.

At the time of the Chernobyl accident, sporadic collection of CO_2 from the atmosphere was initiated at Smara. Three samples were also collected by Göran Possnert, at the Tandem Accelerator Laboratory, Uppsala, on April 29 between 15.00 and 16.00 and 16.00 and 17.00, and on April 30 between 11.00 and 12.00, outside his office window (59°51′N, 18°38′E), and were measured by him, using the AMS technique.

THE EXCESS ^{14}C ACTIVITY FROM AUTUMN 1984 TO AUTUMN 1987

The excess ^{14}C activity is shown in Figure 1. For Abisko, a general decrease of ca 2.5% is to be seen over these three years. There is a tendency to peak between July and August, in 1985 and 1986. The values in 1986 for May and June are almost as low as for April just before the Chernobyl accident. The value for July is, however, the highest during these three years and may have been a real consequence of the accident. The decrease for Svalbard seems to be slightly larger (ca 3.5%), but the general level is also somewhat lower. The pronounced increase for Abisko in the summer after the Chernobyl accident is absent. There may be a normal seasonal increase. Three atmospheric CO_2 samples from Smara indicate higher activity two months after the accident than just after and in the late autumn. Three rhubarb fractions extracted from leaves collected on May 4 give as a weighted mean the excess 211.5 ± 3.4‰, indicating a possible increase at the time of the accident, although the atmospheric sample collected between April 30 and May 4 did not. The three plant samples collected at the end of June and the beginning of July (birch and hazel, respectively) have an insignificantly lower mean excess activity (204.4 ± 3.4‰) than the rhubarb, but a significantly lower activity than the birch plant material (223.8 ± 5.5‰), which assimilated CO_2 from the end of June. The spread between the values within each group (Fig 1) is larger than the uncertainty of the mean value implies. It seems essential that further measurements should be made on the

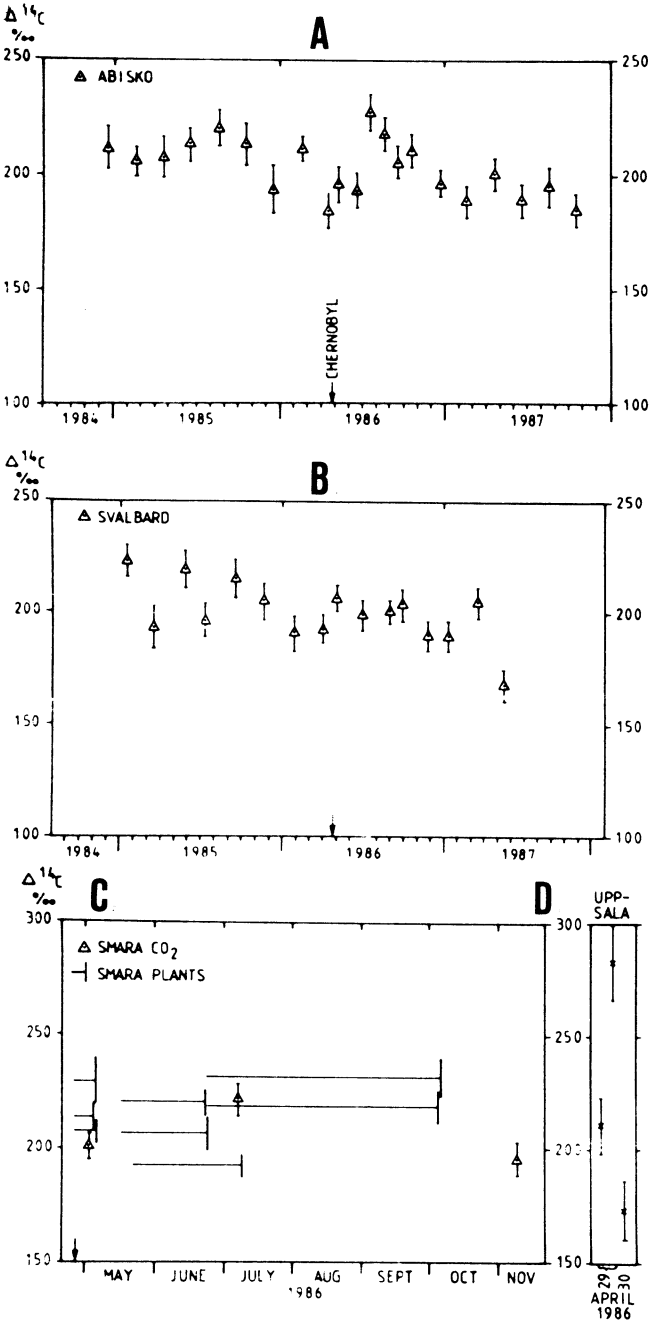

Fig 1. The atmospheric ^{14}C excess from autumn 1984 to autumn 1987 at A. Abisko, Sweden, B. Kapp Linné, Svalbard, C. Smara, Sweden, and D. Uppsala, Sweden. The vertical line indicates when the plant was harvested and the horizontal line, the period during which the plant assimilated CO_2.

remaining plant material. Finally, it will be seen that Possnert's values for Uppsala indicate a significant spread with a high excess for the second sample collected on April 29 and a normal or rather low excess for April 30. It should be recalled that the rainfall and γ-fallout were significant in Uppsala, although negligible at Abisko and Smara in Sweden and Svalbard.

It may be questioned whether the detected variations are real. Internal checks of the statistics were made on the background and standard sample measurements during the period of almost 2.5 years covered by this series of measurements. The ratios of real sigma to expected sigma (σ_R/σ_{exp}) were calculated according to Stuiver (1982). The total period was divided into shorter intervals, because of experiments with the filling pressure, refilling the Geiger counters used in anti-coincidence, and adjustment of the electronics. A summary of these ratios is given in Table 1. Examples of the statistical spread in the form of histograms, together with a diagram showing the slight, steady decrease of the background from February to August 1986 are given by Olsson (1988). All these results indicate that the variations must be regarded as real.

TABLE 1

The statistical quality tests ($\sigma_{real}/\sigma_{expected}$) for background and standard measurements over the last 2.5 years

Sample and period	σ_R/σ_{exp}	No. of measurements
Background		
1986–02–03 – 1986–08–07	0.91	17
1986–08–29 – 1986–10–30	1.44	11
1987–04–21 – 1987–07–06	1.58	10
1987–07–15 – 1987–10–12	0.97	13
1987–12–15 – 1988–05–31	1.32	15
Oxalic acid		
1986–02–05 – 1986–08–15	0.96	14
1986–09–01 – 1986–11–02	1.53	10
1987–03–31 – 1987–08–25	1.04	13
1987–12–15 – 1988–05–31	1.06	18

THE EXCESS ACTIVITY BETWEEN 1978 AND 1982

The use of plant material can be questioned, since plants must have some memory of the nutrients stored in the root system and used in early spring. The sap from birch has been tested and shows some memory: from April 30, 1986, it has a ^{14}C activity corresponding to that of the atmosphere in 1982–83. Similar results are available from earlier measurements. The agreement between activity measured in terrestrial plant material from Smara and elsewhere in Sweden and that in the atmosphere at Abisko is, however, good, as determined from material from the end of 1977 to the beginning of 1983 (Fig 2). The values for the ^{14}C activity of the atmosphere at Kapp

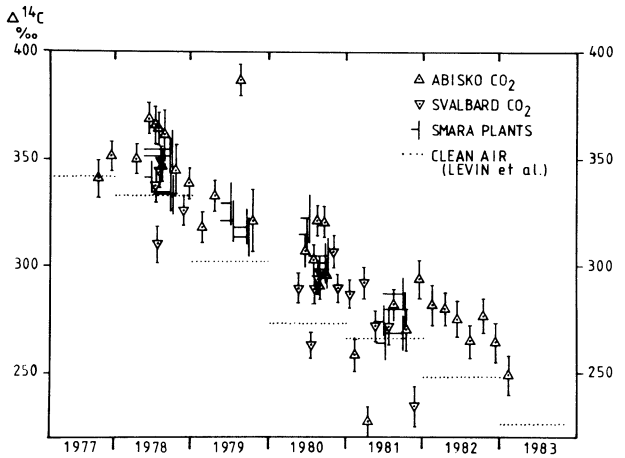

Fig 2. The atmospheric ^{14}C excess from 1978 to 1982 for Abisko, Sweden, and Kapp Linné, Svalbard. Data for various plants are included in the diagram. The vertical line indicates when the plant was harvested and the horizontal line the period during which the plant assimilated CO_2. Single, short, horizontal, dashed lines, indicate "clean" air levels, as given by Levin *et al* (1985).

Linné are included in that diagram, indicating the reality of a generally lower activity at Svalbard than in Sweden. Two plants collected in 1980 at Kung Karls Land, Svalbard, had ca 2% lower excess activity than five plants collected in Sweden. As a comparison, the values for the "clean air" published by Levin *et al* (1985) are included in Figure 2.

SOME LONG-TERM TRENDS

A comparison can be made with values from Nordkapp (Nydal & Lövseth, 1983) which seem to be lower than those for the Abisko samples from 1977 to 1980, for which period many values from the Trondheim and Uppsala laboratories are available. The possibility of a laboratory bias, however, must not be forgotten in such a comparison.

Early results from Uppsala have been published by Olsson and Karlén (1965), Olsson and Klasson (1970), and Stenberg and Olsson (1967) and these also indicate lower activities on Svalbard than in northern Sweden. This could, however, be explained by diffusion to the north and south of ^{14}C entering the troposphere from the stratosphere at northern mid-latitudes and causing the strong seasonal variations observed for many years after 1963. In 1976, when this explanation could hardly have been solely valid, Olsson (1979) proposed a possible island effect in the Arctic region. Plant material from Iceland, Greenland and the Faroes showed slightly lower ^{14}C activities than Swedish plants.

The German values for "clean air" indicate a significantly lower level for the ^{14}C activity than Nordic samples. This must be due to a higher degree of contamination by fossil fuel. Clean air does not exist any longer. The present atmospheric ^{14}C activity at any place is a balance between natural activity, additional anthropogenic activities, dilution by fossil-fuel carbon

dioxide, local natural sources of $^{12}CO_2$, such as volcanoes, and exchange between the reservoirs.

The $\delta^{13}C$ values are changing within a range laid down by a band with a width of ±2‰, containing ca 80% of the measurements, which have gradually become more accurate, as will be seen in the temporal distribution of the outliers. The overall uncertainty has decreased from almost 1‰ to ca 0.3‰. The Abisko values have changed from ca –25‰ in 1961 to ca –26‰ in 1987, and the Svalbard values from ca –25‰ in 1961 to ca –28‰ in 1987. Similar values are obtained even if the values are grouped in four seasons (February–April, etc). An exercise with the $\delta^{13}C$ values for Nordkapp did not yield the same pattern.

DISCUSSION

The rather small decrease of the ^{14}C activity from the end of 1984 to the end of 1987 may have had three causes: 1) the diminishing difference in activity between the atmosphere and the oceans, 2) the Chernobyl Accident counteracting the normal decrease, and 3) some global change in the ocean-atmosphere system affecting the ^{14}C activity.

Since the years 1982 and 1986 exhibit a slower decrease than the general trend, a study of any correlation with the El Niño – Southern Oscillation (ENSO) is urgent. The ENSO events are closely related to the Southern Oscillation Index (giving a measure of the strength of the southeastern trade winds over the northern Pacific), sea-level changes, rainfall, thickening of the pycnocline, changes in the pattern of upwelling water and changes in the sea-surface temperature (Druffel, 1985; Graham & White, 1988). Since El Niño events occur about every fourth year, eg, in 1982 and 1986, it would be of great interest if international cooperation could be established for an intensive study of the ^{14}C activity in the near future, together with the normal variations observed at such events.

CONCLUSION

The decrease of the ^{14}C excess was 1.5 to 2% per year in the last few years before 1985 but ca 1% after that. The "clean" air of Europe has a lower activity than that over most of Sweden (at least north of 59°N). The activity over the Arctic areas, here illustrated by values from Svalbard, is lower than over Sweden. The Chernobyl accident caused much γ-activity fallout in Sweden, well correlated with the rainfall when the wind carrying the released activity swept over part of Sweden. Some increased ^{14}C activity in Sweden later in 1986 and in Uppsala just after the accident may be correlated with the Chernobyl activity. In spite of the general decrease, an earlier period of slightly more than one year (end of 1981 and the whole of 1982) also showed very little decrease. It is too early to draw any conclusion regarding the origin of the detected pattern of the ^{14}C variations. All collected samples for the actual period must be measured and preferably those for yet another year before and after the three-year period now studied in detail.

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