VI. ANTHROPOGENIC ^{14}C VARIATIONS

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ANTHROPOGENIC ^{14}C VARIATIONS*

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ABSTRACT. Continuous ^{14}C data of 1 or 2 weekly samples of atmospheric CO_2 are presented, collected near two nuclear power plants (NPP-Biblis and NPP-Philippsburg) located in the rather densely populated upper Rhine valley. Tree-leaf and tree-ring samples from the area of a boiling water reactor in northern Germany (NPP-Würgassen) are also presented and discussed. Additional atmospheric $^{14}CO_2$ samples from two continental 'clean-air' stations in Germany and Austria were used as reference for the polluted areas. For January 1982, these samples yield a 'clean-air' ^{14}C concentration in central Europe of $\Delta^{14}C=255\pm5‰$[1] (7.7pCi/gC=1.45pCi/m^3 air).

In the vicinity of boiling water reactors we found a mean excess in $\Delta^{14}C$ activity of ca 53% (0.05pCi/m^3 air) above the local level during normal periods of reactor operation. During revision, however, the $\Delta^{14}C$ excess may reach up to 300% above the local background. From this, we calculate source strengths of ca 20Ci/GWa on the average, with peak values of up to 2Ci/week during specific periods. Pressurized water reactors emit ^{14}C mainly as hydrocarbons, and consequently, we found no additional $^{14}CO_2$ near the Biblis reactor.

Stations located in the Rhine valley show significant summer-winter $^{14}CO_2$ variation due to

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* This paper is from an invited talk.

[1] Heidelberg ^{14}C measurements are systematically $\Delta^{14}C=9\pm2‰$ high compared to the NBS Oxalic Acid standard; if to be compared all Heidelberg ^{14}C data should be shifted by $\Delta^{14}C=-9‰$. A careful standard comparison and recalibration is presently under way.

intensive fossil-fuel combustion. The fossil-fuel admixture leads to a depression of the ^{14}C level and reaches $\Delta^{14}C_{depr}$ =120‰ in winter; summer values, however, are only slightly lower than the 'clean-air' level.

INTRODUCTION

In populated areas the 'clean-air' ^{14}C level is modified by two opposite anthropogenic sources:
- fossil fuel CO_2 causes a depletion of the regional ^{14}C level ($^{14}C/^{12}C$ ratio) in industrial areas (Vogel and Uhlitzsch, 1975),
- the emission of ^{14}C from nuclear power and reprocessing plants leads to increased local ^{14}C concentration (Kunz, Mahoney, and Miller, 1974;1975).

The present study continues a previous one by Levin, Münnich, and Weiss (1980). Measurements have been made at the Biblis nuclear power plants (Plants A and B together=2500 MWe) and at the Vermunt and Schauinsland reference stations. A new sampling site was established close to the Philippsburg nuclear power plant (900 MWe). Tree-leaf and tree-ring samples were collected in the area of the Würgassen nuclear power plant (670 MWe). These two plants are boiling water reactors (BWR), emitting mainly $^{14}CO_2$, while pressurized water reactors (PWR) like Biblis primarily emit hydrocarbons (Schwibach, Riedel, and Bretschneider, 1979).

CLEAN-AIR BACKGROUND

'Clean-air' ^{14}C and ^{13}C isotopic measurements are being made at two stations in central Europe, at the Vermunt water power plant (Silvretta 1800m) in Austria (no fossil-fuel contamination, electric power and heating only), and at the UBA-(Federal Environment Agency) observation station on the Schauinsland mountain top (1200m, Black Forest, southern Germany), 1000m above the city of Freiburg.

Figure 1a shows the ^{14}C data for 1976-1982. After the nuclear weapon test ban in 1963, the ^{14}C level decreased exponentially ($\tau \approx 15$yr), due to the exchange between atmosphere and ocean (see Levin, Münnich, and Weiss, 1980, Fig 1).

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For the last few years, a linear regression fits to the observed data equally well:

$$\Delta^{14}C = 375 - 20 * t^2 \quad t = \text{years since } 1/1/76 \quad (1)$$

The fact that Vermunt and Schauinsland data agree well indicates that the data of both stations may be used as ^{14}C reference on the continent, not influenced locally, but rather representing the general ^{14}C level in central Europe. The summer-winter peak-to-peak variations up to $\Delta^{14}C$=40‰ are essentially due to a generally higher pollution level of the continental air during winter: The corresponding $\delta^{13}C$ values indicate that the Schauinsland station, although only 1000m above Freiburg and the Rhine valley measures the local pollution level sufficiently low except for a remaining contribution of about 4 to 5 ppm fossil-fuel CO_2 ($\Delta^{14}C_{depr}$=15‰, $\Delta^{14}C_{depr}$ = depression below the 'clean-air' level) in winter. The remaining long-term deviation, especially during 1978/79 when the 'clean-air' ^{14}C level was ca $\Delta^{14}C$=14‰ above the linear fit shows no significant seasonal variation. It presumably results from post-test-ban atmospheric nuclear weapon tests in China (9/17/77, 3/15/78, 12/14/78) (Fry et al, 1981, Levin, Münnich, and Weiss, 1980).

ATMOSPHERIC SAMPLES FROM BIBLIS AND HEIDELBERG STATIONS

Both sampling locations, Biblis and Heidelberg, are situated in the Rhine valley, a region rather densely and evenly populated. Figure 2 shows their locations and the regional fossil-fuel pollutant sources in the Rhine valley, i e , primarily the cities of Mannheim, Ludwigshafen (industrial area), Worms, and Speyer.

Levin, Münnich, and Weiss (1980) found that the nuclear power plant Biblis A, a pressurized water reactor, is not a source for $^{14}CO_2$. Figure 1b shows the time variations of the $^{14}CO_2$ concentration in Biblis where the ^{14}C level is obviously only influenced by fossil-fuel CO_2. The mean depression during the winter half of the year is ca $\Delta^{14}C_{depr}$=60‰ with peak depressions up to 100‰, while the summer values are only slightly lower than the 'clean-air'

[2] See footnote 1

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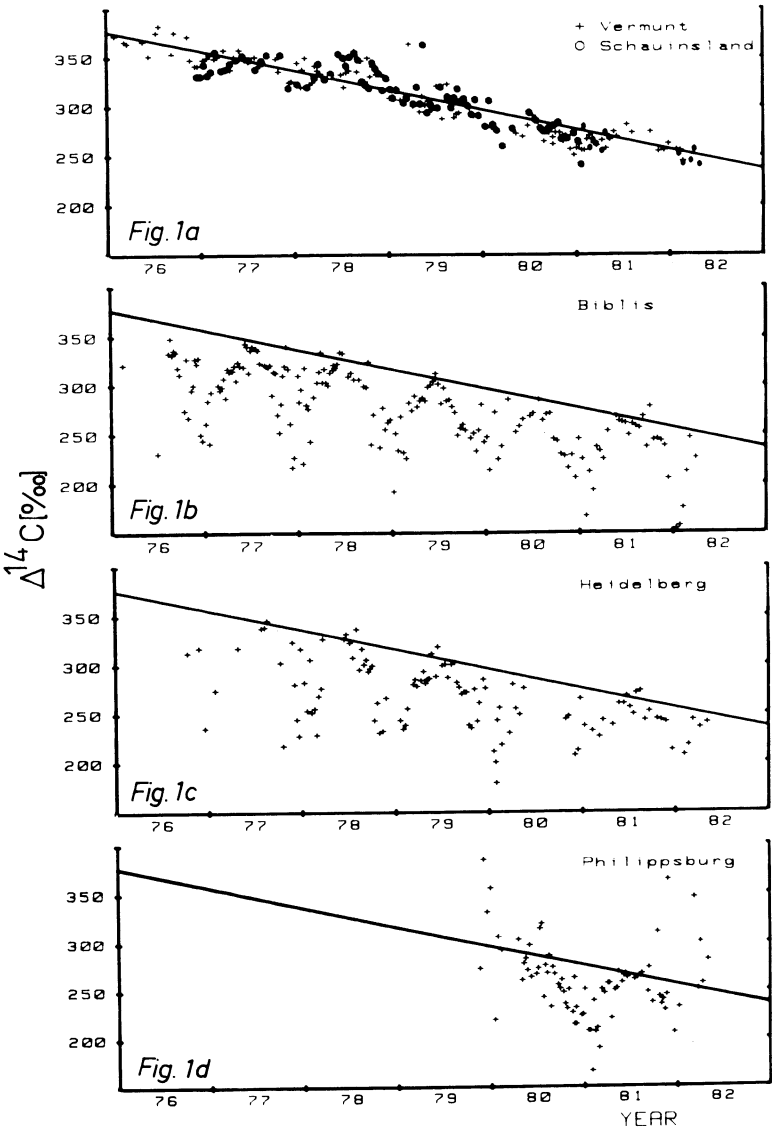

Fig 1. ^{14}C concentration of atmospheric CO_2 at Vermunt and Schauinsland reference stations, 1a. At NPP-Biblis in the Rhine valley, 1b. At the Heidelberg institute ca 15m above the ground, 1c. At sampling site ca 1.5 km northwest of the NPP-Philippsburg, 1d. (See map, Fig 2). The straight line is a least squares fit through the reference station data.

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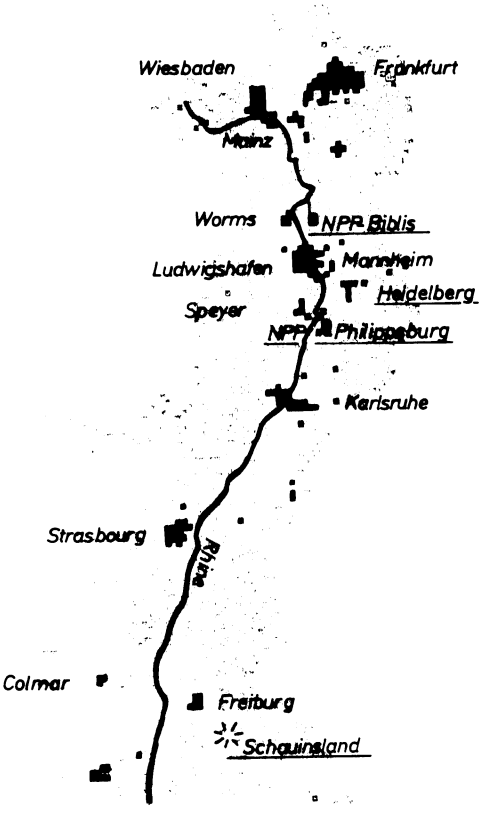

Fig 2. Sources of ^{14}C-free CO_2 from fossil-fuel combustion in the Rhine valley; shaded areas represent the homogeneously distributed smaller anthropogenic sources like villages, traffic, etc. Black areas indicate cities. Underlined names stand for sampling sites (modified from Bartholomäi and Kinzelbach, 1980).

reference level. The same winter depression is observed at the Environmental Physics Institute (Fig 1c), located on the outskirts of Heidelberg. Similar summer-winter variations were observed by Tans (1978) on the Smilde Drenthe radio tower station 100m above the ground (Netherlands). Tans interprets part of these variations as the result of pulsed stratospheric injections of natural and bomb ^{14}C. Unfortunately, there is only one year overlap with our Biblis and Heidelberg data. From the little corresponding 'clean-air' data that we have from Vermunt for 1975/76 we cannot distinguish any significant summer-winter variation. But as indicated before the slight seasonal ^{14}C variations in the period from 1977 to 1981 have no significant contribution from stratospheric ^{14}C injections.

Moreover, the summer maximum observed by Tans

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lies on the reference fit[3] of the mountain stations of southern Germany and Austria. The low winter values observed by Tans are in phase with those of Biblis and Heidelberg, but the amplitude is only ca 40% of the one in the Rhine valley. If the Tans minima are in fact due to fossil-fuel CO_2, the reduced amplitude might roughly represent the time fraction during which air masses originate from land, while the rest of the time, unpolluted air comes from the ocean.

ATMOSPHERIC SAMPLES FROM THE PHILIPPSBURG BOILING WATER REACTOR

The Philippsburg nuclear power plant is a BWR of about the same power production as each of the Biblis reactors. It is located at the Rhine river south of Mannheim and Ludwigshafen. Spot measurements in the reactor stacks of BWRs yielded source rates of 5-16Ci/GWa of ^{14}C emitted as $^{14}CO_2$, PWRs emit hydrocarbons at a rate of 3-11 Ci/GWa (Schwibach, Riedel, and Bretschneider, 1979).

The Philippsburg reactor stopped in May 1980, resuming operation in October 1981. It is now inoperative since May 1982. In 1980/81 we observed the same winter depression as in Biblis and Heidelberg. In spring 1980 and winter 1981/82 $^{14}CO_2$ levels were observed significantly above the reference line (see Fig. 1d). From November 1979 to May 1980 we found $\Delta^{14}C$ to be 10 to 130‰ higher than the level in the Rhine valley which is uninfluenced by the nuclear power plant. The mean value is $\Delta^{14}C=296\pm5‰$[1], that is 24‰ above the Rhine valley level and 14‰ above the 'clean-air' level. The highest ^{14}C activities (eg. $\Delta^{14}C=570‰$[1] in October 1981) occurred a short time before or a few days after operation. Perhaps the containment was flushed during these periods.

Using a Gaussian plume model with dispersion parameters of Vogt (1970), and the actual synoptic data from the Mannheim Weather Service station (Deutscher Wetterdienst, 1979-1982) we calculated dispersion factors at the sampling location for the periods when the reactor was running. Together with

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[3] See footnote 1

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the excessive $\Delta^{14}C$ levels of the two periods above, we have estimated the ^{14}C source strengths. The mean source strength for November 1979 to May 1980 was 16Ci/yr and 18Ci/yr for October 1981 to May 1982. The samples with highest ^{14}C levels indicated source strengths of up to 2Ci/week (≃100Ci/yr) during the sampling period. There are no published direct measurements of the source strength from stack samples of the Philippsburg reactor. Our results, however, fit the average source strength of boiling water reactors in western Germany quite well.

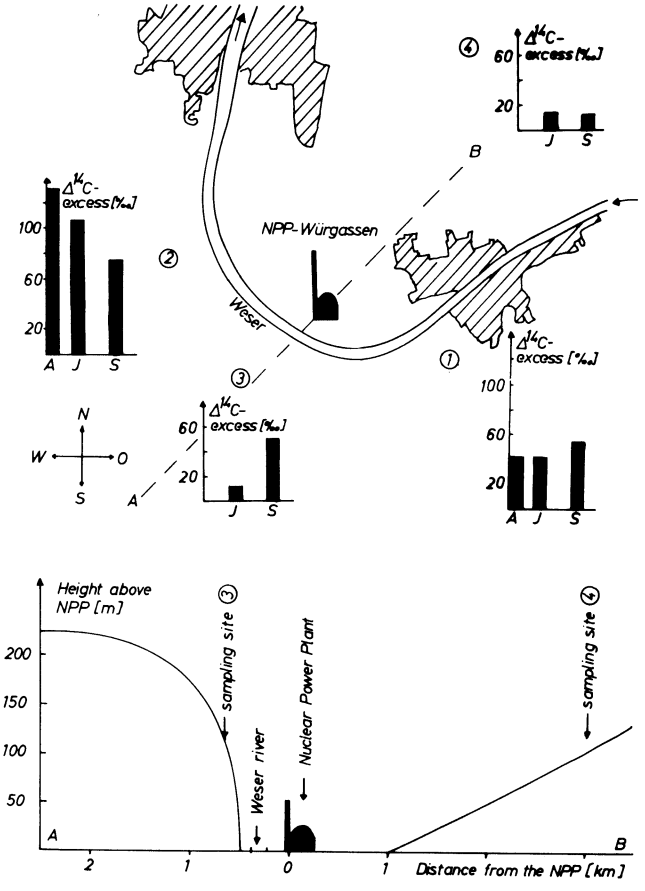

Fig 3. Top:geographic location map of the Würgassen NPP in the Weser valley. Diagrams 1-4 represent the observed $\Delta^{14}C$ excess over the 'clean-air' level in tree leaves collected at different times of 1981 at the corresponding sites (1-4). Bottom: topographic profile along line A - B (see top) illustrating hilly terrain.

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PLANT SAMPLES FROM THE WÜRGASSEN NUCLEAR POWER PLANT

Würgassen is a BWR of 670MWe, in the Weser River valley of the northern part of western Germany near Kassel and Göttingen. Tree-leaf samples were collected at four sites around the reactor (Fig 3) in April, June, and September 1981; tree-ring samples were taken from trees at Sites 1 and 4. The greatest ^{14}C effects were expected at Site 1, the lowest at Site 4, because of the mean geostrophical wind direction from the west, and because ground-level wind is canalized by the valley (hills in the south and west of the river reach heights up to 250m). Atypically, there was a very stable weather pattern in April 1981, with constant winds from the east causing a strong ^{14}C increase in the leaves collected at Site 2 ($\Delta^{14}C_{excess}$=130‰, $\Delta^{14}C_{excess}$ = positive deviation from the 'clean-air' level). Leaves collected at Site 1 show only little excess, $\Delta^{14}C_{excess}$=40‰(Fig 3). Although the situation changed

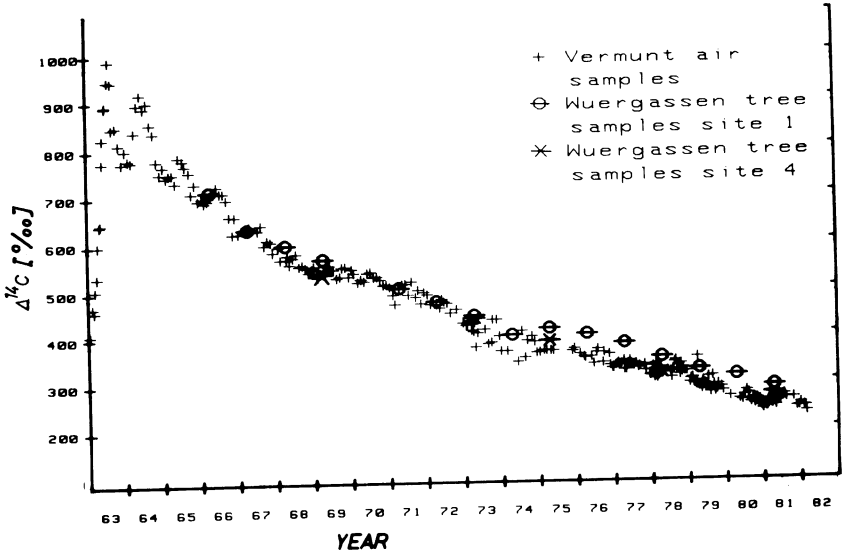

Fig 4. ^{14}C concentration of atmospheric CO_2 at Vermunt together with the tree-ring samples from trees felled near Würgassen NPP at Sites 1 and 4 (see map Fig 3).

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due to 'normal' weather conditions with wind from the northwest, all tree leaves collected in the course of the vegetation period 1981 (in June and September) show only slight changes in ^{14}C content (decrease at Site 2, increase at Site 1).

Figure 4 shows the results of tree-ring measurements from Sites 1 and 4. The reactor has been operative since 1972. During this time, the tree-rings of Site 1 are generally ca 35‰ higher in $\Delta^{14}C$ than at Site 4, which shows nearly background concentrations (mean $\Delta^{14}C_{excess}$=10‰).

Dispersion models are not as yet applicable to hilly terrain which prevents the calculation of source strengths from observed ^{14}C data. Nevertheless, our measurements do not indicate higher source strengths than expected for older type German BWRs (up to 20Ci/GWa).

COMPARISON OF INTERHEMISPHERIC MIXING DATA AS DERIVED FROM BOMB ^{14}C AND FROM ^{85}Kr

On the basis of nuclear test ^{14}C data Münnich and Vogel (1963) used a global diffusion model for deriving a relaxation time of ca 5 months for the lowest order sperical harmonic representing an interhemispheric atmospheric concentration difference. This corresponds to a hemispheric residence time of 10 months. Czeplak and Junge (1974) used a similar, but more sophisticated model and also reviewed all available data of other tracers. They concluded that ca 1 year was the most probable value for the hemispheric residence time.

Until recently, nuclear bomb ^{14}C provided the most reliable basis for estimating interhemispheric exchange. Since very accurate ^{85}Kr data are available (Weiss et al, in press) it is interesting to compare the respective results since bomb ^{14}C has its source primarily in the stratosphere while ^{85}Kr originates from fuel reprocessing at ground level. The meridional ^{85}Kr profiles show a significant concentration drop at the Intertropical Convergence, suggesting a slightly longer hemispheric residence time. However, the difference is not positively significant.

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