

## THE EFFECT OF ANTHROPOGENIC CO<sub>2</sub> AND <sup>14</sup>C SOURCES ON THE DISTRIBUTION OF <sup>14</sup>C IN THE ATMOSPHERE

INGEBORG LEVIN, K O MÜNNICH, and WOLFGANG WEISS

Institut für Umweltphysik, Universität Heidelberg,  
Federal Republic of Germany

**ABSTRACT.** <sup>14</sup>C measurements on continuous weekly samples of atmospheric CO<sub>2</sub> and hydrocarbons, collected in a rather densely populated area are presented. The deviation of the measured <sup>14</sup>C data from the clean air level is primarily due to CO<sub>2</sub> from the combustion of fossil fuels. This is confirmed by fossil fuel admixture estimates individually calculated with an atmospheric dispersion model. Up to 10 percent admixture is predicted by this model and observed from the <sup>14</sup>C shift for weekly averages, particularly during the winter season. Natural CO<sub>2</sub> admixture due to soil respiration, however, even in winter, is of the same order of magnitude, but much larger in the warm season: the considerable variations in CO<sub>2</sub> concentration in summer are almost exclusively controlled by natural sources. Using tree leaf samples, we have been able to identify boiling water reactors (BWR) as weak sources of <sup>14</sup>CO<sub>2</sub>. Atmospheric samples taken in the environment of the pressurized water reactors (PWR) Biblis show that the <sup>14</sup>C release of these reactors is primarily in the form of hydrocarbon <sup>14</sup>C. The source strength of the various power plants, calculated on the basis of our observations in their environment, ranges from 0.5 to 7Ci per year.

### INTRODUCTION

The clean air <sup>14</sup>C level is modified in densely populated areas by anthropogenic sources: 1) fossil fuel CO<sub>2</sub> (<sup>14</sup>C free) causes a depletion of the regional <sup>14</sup>C level in industrialized areas (Vogel and Uhlitzsch, 1975). 2) The emission of <sup>14</sup>C from nuclear power plants (<sup>14</sup>CO<sub>2</sub> mainly from boiling water reactors, radioactive hydrocarbons from pressurized water reactors [Kunz, Mahoney, and Miller, 1974; 1975]) leads to an increase of the local <sup>14</sup>C concentrations.

A study has been made in the Rhine Neckar area (Mannheim/Ludwigshafen/Heidelberg) to distinguish between the influence of fossil fuel combustion CO<sub>2</sub> from the industrialized area around Mannheim and the <sup>14</sup>C emission from the Biblis nuclear power plants.

In addition to this study, based on direct atmospheric CO<sub>2</sub> and hydrocarbon measurements, we collected tree-leaf and tree-ring samples in the vicinity of a number of German power plants to monitor any <sup>14</sup>CO<sub>2</sub> releases from the nuclear reactors.

#### *Clean air <sup>14</sup>C background*

The identification of the different anthropogenic CO<sub>2</sub> sources by isotopic analysis requires knowledge of the clean air isotopic composition. From 1959 (beginning of nuclear weapon testing) to the present, we have made atmospheric CO<sub>2</sub> measurements at several clean air stations in Western Europe and South Africa (Berdau and Münnich, 1972; Vogel and Marais, 1971) and since 1972, primarily at the stations, Vermunt, Vorarlberg, Austria, 1800m, and Schauinsland, near Freiburg, West Germany, 1284m. The time variation is shown in figure 1. An exponential regression, due to exchange between atmosphere and ocean (Münnich, 1963), of the data points gives a clean air <sup>14</sup>C concentration in January 1979 of  $\Delta^{14}\text{C} = 323 \pm 1\text{‰}$ . The fitted curve was used as clean air back-

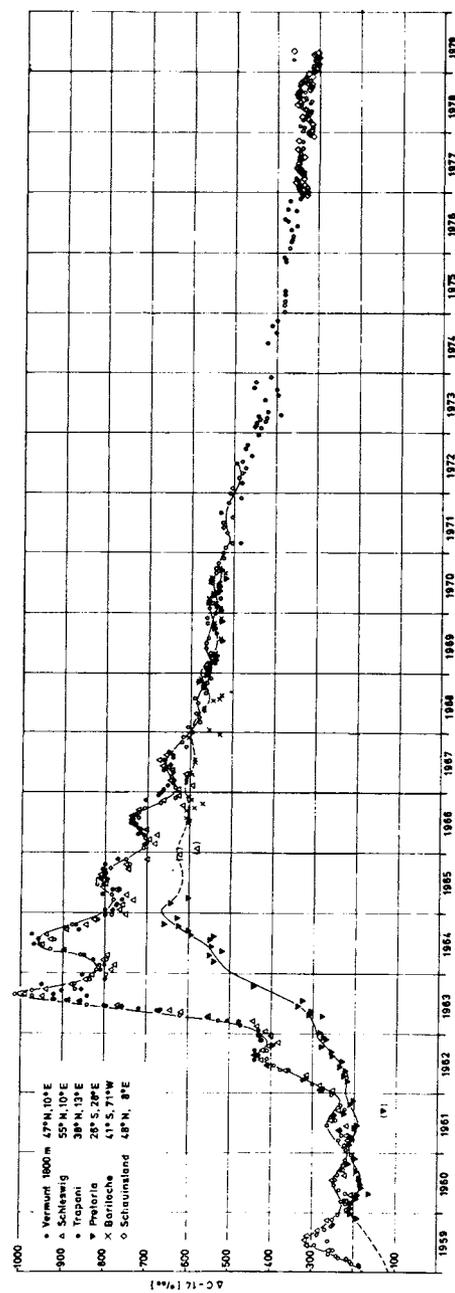


Fig 1.  $^{14}\text{C}$  concentration of tropospheric  $\text{CO}_2$  since 1959; the lower curve represents the  $^{14}\text{C}$  concentration in the southern hemisphere (Pretoria and Bariloche, South Africa) and the upper one shows the time variation in the northern hemisphere (Trapani, Schleswig, Vermunt and Schainzland, Western Europe) (Münich and others, ms in preparation).

ground for the atmospheric samples from 1976 to 1977. The strong variation,  $\Delta^{14}\text{C}$  maximum in early summer, observed in clean air in 1978, possibly resulting from later atmospheric nuclear weapon tests (Chinese) and induced by the exchange between troposphere and stratosphere (Münnich, 1963), were taken into account by a separate fit of the clean air values for 1978.

### Sampling methods

Since August 1976, continuously collected weekly samples of atmospheric CO<sub>2</sub> and total carbon (CO<sub>2</sub> plus oxidized hydrocarbons) were taken about 4 km east of the nuclear power plants in Biblis (Levin, 1978). The sampling system is shown schematically in figure 2. About 15 m<sup>3</sup> of air are sampled by a small aquarium pump. The exact volume of air is measured by a gas meter. The air then passes through a glass column filled with 4n sodium hydroxide solution. To strip the air quantitatively from CO<sub>2</sub>, the glass column contains a packing of small Raschigrings (hardglass) to enlarge the absorption surface. The column rotates around its axis causing permanent renewal of the NaOH film on the column packing. Parallel to this, in a second path, the atmospheric hydrocarbons are oxidized catalytically at 600°C to CO<sub>2</sub> and are then absorbed together with the atmospheric CO<sub>2</sub> in an absorption system identical to the one described before.

The glass columns are changed once a week. The absorbed CO<sub>2</sub> sample is extracted directly in the sampling device in a laboratory vacuum system (fig 3) by addition of half-concentrated H<sub>2</sub>SO<sub>4</sub> (40 percent) to the sodium carbonate solution. The CO<sub>2</sub> samples are cleaned in a charcoal column and measured in a CO<sub>2</sub> proportional counter (*cf* Schoch and others, 1980).

The tree-leaf samples, mainly from old fruit trees, standing rather isolated, were collected in 1976, 1977 and 1978 at the German reactors Biblis (PWR, 2500 MWe), Brunsbüttel (BWR, 806 MWe), Gundremmingen (BWR, 252 MWe), Karlsruhe (PWR, 57 MWe), Lingen (BWR,

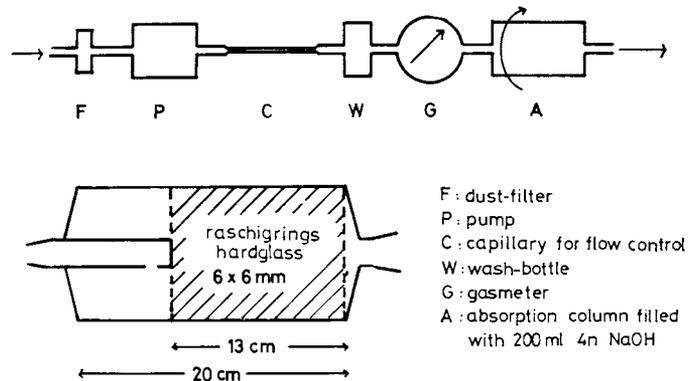


Fig 2. Absorption system for atmospheric CO<sub>2</sub> samples with a rotating NaOH absorption column.



the sampling station. Two other large sources in the vicinity of the sampling site are Darmstadt and Mainz.

In order to distinguish between the influence of the reactor and of the combustion sources which compensate each other to a certain extent, we first estimated the fossil fuel admixture at the sampling point with an atmospheric dispersion model. The model is based on a circular area source for fossil  $\text{CO}_2$  (fig 5) around the sampling point with a radius of 40km. All sources outside this area are assumed not to contribute to the depression at the sampling point. The circular area is cut off in the east and west due to the specific topography of the Rhine valley. Sources on the hills, about 300m above the valley, presumably do not contribute due to a larger vertical dispersion. The source strength has been assumed to be proportional to the population density. This assumption has been checked with the sales statistics of fossil fuel for individual areas: average consumption is presumably 3t of carbon per person per year (Statistisches Landesamt Baden-Württemberg, 1976). This leads to a mean area source strength in the model area (500 inhabitants per  $\text{km}^2$ ) of 1500t carbon per  $\text{km}^2$  per year; assumed seasonal variation: summer, 75 percent, winter, 125 percent. Big cities such as Mannheim/Ludwigshafen, Darmstadt, and Mainz are considered as additional sources (see fig 5). Together with three-hour synoptical data, wind direction and velocity, cloudiness, solar radiation (Deutscher Wetterdienst, 1976-1979), we calculated the theoret-

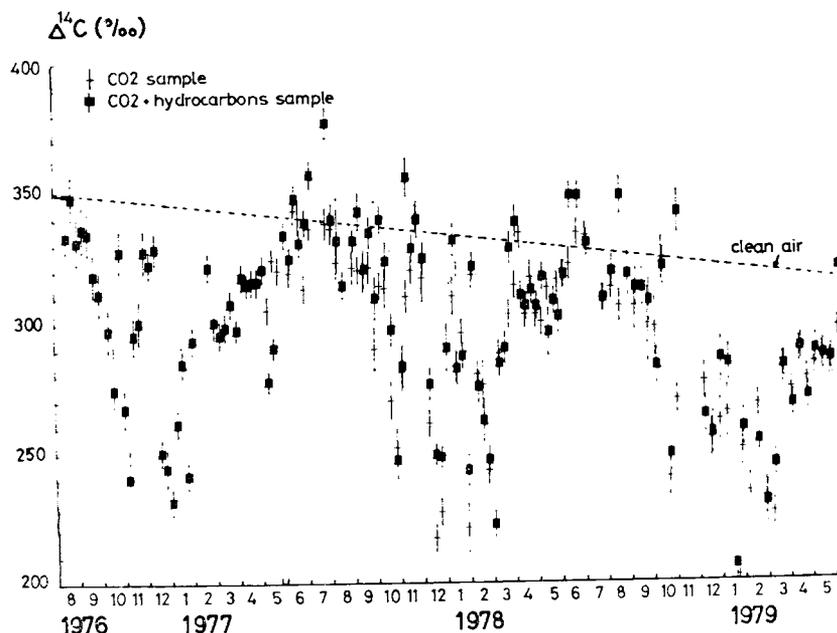


Fig 4. Time variation of  $^{14}\text{C}$  in atmospheric  $\text{CO}_2$  (+) and  $\text{CO}_2$  + hydrocarbons (■) samples, collected near the nuclear plants in Biblis (FRG). The dotted line indicates the interpolated clean air  $^{14}\text{C}$  level.

ical fossil fuel admixture at the sampling location for each weekly sample. Depending on individual wind direction and speed, air has varying durations over the source area and carries varying concentrations of fossil  $\text{CO}_2$  on arriving at the sampling site. The fossil fuel concentration observed at the sampling point further depends on actual atmospheric mixing conditions (stability) controlling vertical dilution of the source  $\text{CO}_2$  (Klug, 1964; 1969).

The results of the model calculation are shown in figure 6. The correlation between the estimated  $\text{CO}_2$  admixture and the one observed is quite good ( $\chi^2 = 0.68$ , [Bevington, 1969]). Assuming that the sample contains only two components, clean air  $\text{CO}_2$  and fossil fuel  $\text{CO}_2$ , neglecting, for the time being, any reactor influence (the Biblis reactors are of pressurized water type and emit  $^{14}\text{C}$  primarily in reduced form) and assuming (cf Dörr and Münnich, 1980) that, at present,  $^{14}\text{C}$  in soil-born  $\text{CO}_2$  is not significantly different from the atmosphere, the fossil fuel mixing ratio can be calculated from the  $\Delta^{14}\text{C}$  value observed, compared to the clean air concentration from the following equation:

$$\frac{C_{\text{fossil fuel}}}{C_{\text{clean air}}} = \frac{\Delta^{14}\text{C}_{\text{clean air}} - \Delta^{14}\text{C}_{\text{sample}}}{\Delta^{14}\text{C}_{\text{sample}} + 1000} (\%) \quad (1)$$

The linear regression in figure 6 does not show a significant reactor influence that would cause a decrease of the measured  $^{14}\text{C}$  depression

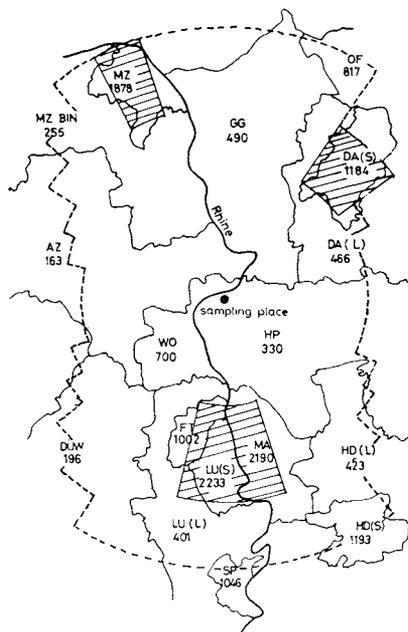


Fig 5. Map of the area source of fossil fuel combustion  $\text{CO}_2$  in the Rhine valley around the sampling site near the Biblis reactor. The map shows the mean population density (number of inhabitants per  $\text{km}^2$ ) of the various districts.

(the regression line passes, in fact, through the origin). This finding agrees well with results of spot measurements made directly in the reactor stacks (Riedel and Gesewsky, 1977). The points in brackets seem to indicate short-term <sup>14</sup>C releases from the reactor. They were sampled between 16.5 and 21.5.78, and between 29.5 and 8.6.78. Indeed, there has been a maintenance period of the Biblis reactor B during June 1978, so that a <sup>14</sup>C release from the reactor might be possible. In these two specific cases the appropriateness of the depression estimated by the model is also supported by the observed <sup>13</sup>C values. A fossil fuel, but likewise a soil CO<sub>2</sub> admixture changes the δ<sup>13</sup>C due to the different (<sup>13</sup>C)/(<sup>12</sup>C) ratios of fossil fuel and plant material in general (δ<sup>13</sup>C = -25‰) and of global atmospheric CO<sub>2</sub> (δ<sup>13</sup>C = -8‰). Thus, observed δ<sup>13</sup>C values of one week's integrated samples from low altitude locations vary in the range of -8 to -11‰ in good correlation with individual CO<sub>2</sub> concentration (Esser, 1975). Note that the slope of the regression line in figure 6 is not exactly equal to 1. This might be due to a source strength assumed too high in our calculation.

2) The higher Δ<sup>14</sup>C values observed for oxidized samples in Biblis can only result from reactor-released <sup>14</sup>C in reduced form. The observed effect could not result from natural methane because mean Δ<sup>14</sup>C methane values much greater (about 1000‰) than ever observed were necessary for

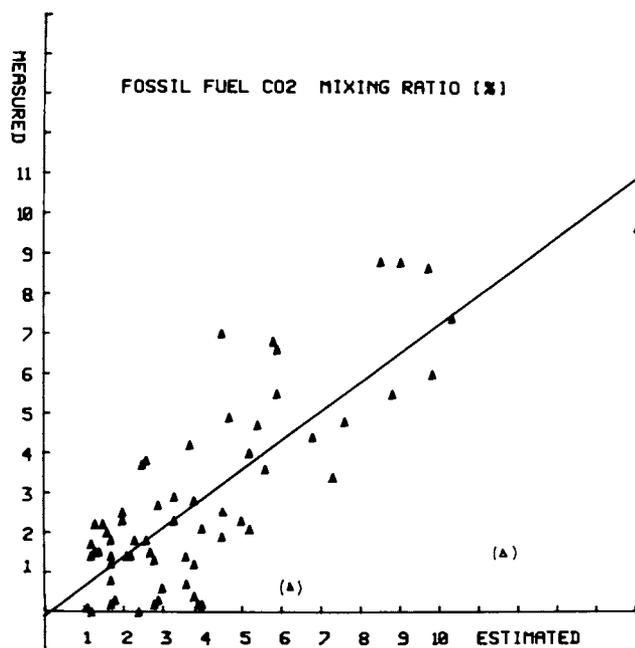


Fig 6. Correlation between the measured fossil fuel CO<sub>2</sub> admixture (calculated from the observed <sup>14</sup>C depression in the samples) and the CO<sub>2</sub> admixture estimated by the dispersion model.

the increase in the composite sample. Atmospheric methane indeed shows  $\Delta^{14}\text{C}$  values of about 0‰ (Levin, 1978).  $^{14}\text{C}$  excess of the hydrocarbons was calculated by subtraction from the  $\Delta^{14}\text{C}$  values of the  $\text{CO}_2$  samples simultaneously collected. Estimated excess of the oxidized samples is shown in figure 7. The mean value over the whole period is  $\Delta^{14}\text{C} = + (8.3 \pm 1.4)\text{‰}$  corresponding to an excess of 6‰ relative to the clean air  $^{14}\text{CO}_2$  concentration. With a long-term dispersion factor of  $f = 8.2 \cdot 10^{-8} \text{ s/m}^3$  for the sampling point, with respect to a point source represented by the Biblis reactors, this increase leads to a source strength of 3.5Ci per year.

*Measurements of plant material near several German power plants*

Interpretation of tree-leaf and tree-ring samples requires knowledge of the exact growth ( $\text{CO}_2$  collection) period of the plant material to enable us to account for the proper clean air  $^{14}\text{C}$  level. For this purpose, we defined general "sample periods" for tree leaves — beginning of April to end of May — and for wood — beginning of April to end of October — (Becker, 1978, pers commun). For these periods, we calculated the clean air reference value as a mean of the observed clean air  $^{14}\text{C}$  concentrations:

$$1976 : \Delta^{14}\text{C} = 368 \pm 3\text{‰}$$

$$1977 : \Delta^{14}\text{C} = 347 \pm 1\text{‰}$$

$$1978 : \Delta^{14}\text{C} = 336 \pm 1\text{‰}$$

Significant increases above these levels were only found in leaf samples

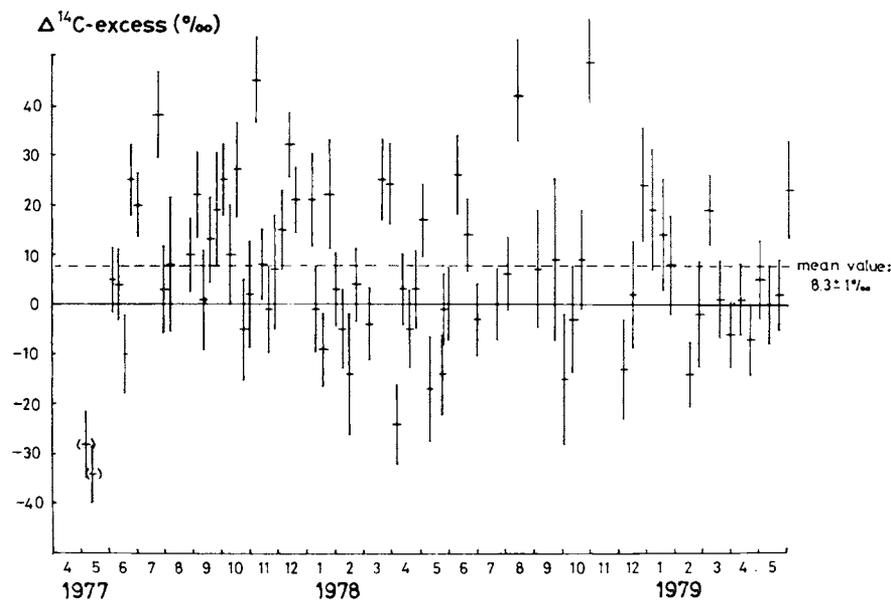


Fig 7.  $\Delta^{14}\text{C}$  excess of the combined  $\text{CO}_2$  + hydrocarbons samples compared to  $\text{CO}_2$  samples collected near Biblis reactors. The mean excess is  $\Delta^{14}\text{C} = + (8.3 \pm 1.4\text{‰})$

collected near the Würiggassen and Obrigheim reactors in 1976 and near the Brunsbüttel reactor in 1977:

$$\text{mean } \Delta^{14}C_{\text{increase}} \text{ (Obrigheim 1976)} = 19 \pm 2\text{‰}$$

$$\text{mean } \Delta^{14}C_{\text{increase}} \text{ (Würiggassen 1976)} = 17 \pm 2\text{‰}$$

$$\text{mean } \Delta^{14}C_{\text{increase}} \text{ (Brunsbüttel 1977)} = 19 \pm 2\text{‰}$$

These values correspond to a  $^{14}C$  excess of about 2 percent of the natural  $^{14}C$  concentration.

The results of the Obrigheim tree-ring measurements are plotted in figure 8. In accordance with the tree-leaf data, we found an average excess in the concentration of wood of  $\Delta^{14}C = 27 \pm 3\text{‰}$  over the whole period, 1968 to 1978. Particularly in the years 1970, 1974, 1975, and 1978, we observed a significant excess of  $\Delta^{14}C = 35 \pm 9\text{‰}$  to  $49 \pm 9\text{‰}$ .

To calculate  $^{14}C$  source strengths from the observed excess, a record of the actual atmospheric dispersion conditions during the growth period is needed. As such a record was not available to us, we tried to estimate the releases according to equation (2) using long-term dispersion factors,  $f$ , which describe the mean yearly dispersion conditions for the different sampling locations. Data was provided by H Riedel, Neuherrberg, West Germany (1977, pers commun). The dispersion factors are calculated from long-term synoptical data records at the nuclear power plant sites.

$$Q = c_+ / f \text{ [Ci/s]} \quad \begin{array}{l} Q : \text{source strength [Ci/sec]} \\ f : \text{long-term dispersion factor [s/m}^3\text{]} \\ c_+ : \text{additional } ^{14}\text{C concentration [Ci/m}^3\text{]} \end{array} \quad (2)$$

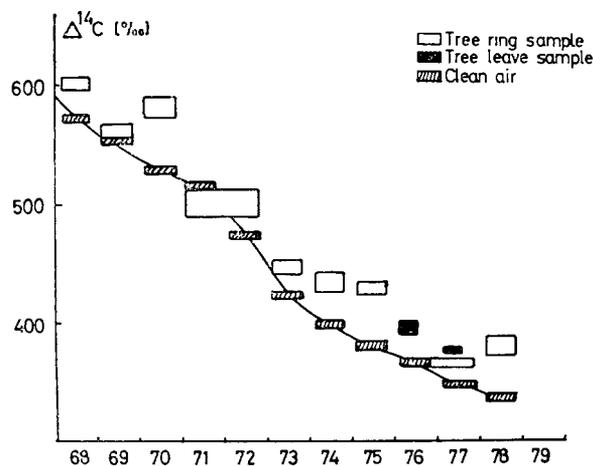


Fig 8. Measured  $^{14}C$  concentrations of tree-ring samples collected near Obrigheim reactor ( $\square$ ) and tree leaf samples from Obrigheim, Würiggassen, and Brunsbüttel reactor ( $\blacksquare$ ) compared with the mean clean air level ( $\text{hatched}$ ) during the growth period.

We estimated  $^{14}\text{C}$  emissions of about 0.5 to 7Ci per year per nuclear power plant, which again agree well with direct emission measurements in the power plant stacks (Riedel and Gesewsky, 1977). Nevertheless, those estimates can only give the order of magnitude of the real  $^{14}\text{C}$  emission because the long-term dispersion factors describe the mean dispersion condition during the whole year, which may differ considerably from the actual situation, especially if the  $^{14}\text{C}$  release is not constant with time.

Tree leaves sampled around the Biblis reactors showed no  $^{14}\text{C}$  increase above the clean-air level. They show a little but significant  $^{14}\text{C}$  depression due to fossil fuel admixture. This behavior is similar to that found in  $\text{CO}_2$  samples collected directly from the atmosphere, as discussed above. The  $^{14}\text{C}$  depression in plant material is, however, much smaller, with an average reduction in  $\Delta^{14}\text{C}$  of only  $-(5 \pm 1)\text{‰}$ , than it is in atmospheric  $\text{CO}_2$  sampled during the same season, eg, spring. Reduction in  $\Delta^{14}\text{C}$  in atmospheric  $\text{CO}_2$  amounts to  $-(24 \pm 1)\text{‰}$ , on the average. This remarkable difference is due to the fact that  $\text{CO}_2$  samples were collected all day, while plants, of course, only sample during the daytime when atmospheric stability is much lower and mixing is enhanced. In fact, our atmospheric mixing model correctly predicts varying fossil fuel contributions to plant material if it runs between sunrise and sunset only (Levin, 1978).

#### CONCLUSION

The data shows that reactor-produced radiocarbon is detectable in its environment, although the concentration is rather low in the cases

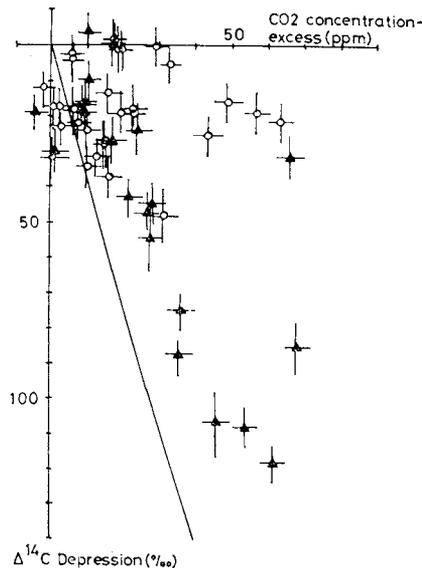


Fig 9.  $\Delta^{14}\text{C}$  depression versus  $\text{CO}_2$  concentration excess: the line represents the regression of  $\text{CO}_2$  concentration excess and  $\Delta^{14}\text{C}$  depression if caused by exclusive admixture of fossil fuel  $\text{CO}_2$ . The deviation from this line is due to admixture of soil respiration  $\text{CO}_2$  (summer ( $\circ$ ), winter ( $\blacktriangle$ ) samples.)

studied here. Air concentrations due to both reactor <sup>14</sup>C emissions as well as fossil fuel CO<sub>2</sub> emissions, strongly depend on the actual meteorological situation, primarily atmospheric stability. Even in a rather densely populated region, however, the source flux densities due to natural sources and sinks, soil respiration (see *cf* Dörr and Münnich, 1980) and plant assimilation during the daytime, are considerably greater than man-made ones, typically 25 to 50g CO<sub>2</sub>/m<sup>2</sup> • day as compared with, for example, 15g/m<sup>2</sup> • day due to fossil fuel in the Rhine-Neckar region study in this paper.

This becomes evident from figure 9 where the individual <sup>14</sup>C depression is plotted versus the observed excess in CO<sub>2</sub> concentration. Nearly all data points show a significantly larger CO<sub>2</sub> concentration excess than can be caused by the admixture of fossil fuel CO<sub>2</sub> (definitely known from the <sup>14</sup>C depression observed). This deviation from the fossil fuel regression line is due to admixture of plant and soil respiration CO<sub>2</sub>. The interpretation is unambiguous since this CO<sub>2</sub> is not significantly different in <sup>14</sup>C from the normal atmospheric CO<sub>2</sub> (*cf* Dörr and Münnich, 1980). Thus, admixture of respiration CO<sub>2</sub> shifts the point into positive x direction only. One sees that this primarily occurs in summer when the admixture of natural CO<sub>2</sub> may be up to five times the fossil fuel one. The considerable variations in atmospheric CO<sub>2</sub> concentration observed (a range of about 30ppm, *ie*, 10 percent of the average atmospheric concentration, during the same day is often found in the summer even in a flat unforested area about 20m above ground) are primarily due to natural sources even in densely populated areas. Therefore, we now try to model these variations in a similar way to the one reported here. In this context, a simultaneous study of atmospheric CO<sub>2</sub> and atmospheric radon-222 seems to be very helpful.

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## DISCUSSION

*Loosli*: You estimate admixtures of fossil fuel  $\text{CO}_2$  to your weekly samples by using an atmospheric dispersion model. How precise are these estimated values and which sources are included in this error?

*Levin*: The coefficient of the calculated regression line from the correlation between model estimated and measured admixtures show an error of at least 30 percent. Due to the uncertainty in the synoptical data and the resulting diffusion parameters, an appreciably larger error (factor of 2) should be expected.

*Haas*: The soil gases reflect the type of vegetation growing in the area, *ie*, C-3 or C-4 type plants. In the USA, a range of  $\delta^{13}\text{C}$  from  $-17\text{‰}$  to  $-26\text{‰}$  was observed according to plant types growing in Texas or North Dakota, respectively.

*Levin*: Our own measurements from this region show  $\delta^{13}\text{C}$  values of soil respiration  $\text{CO}_2$  of about  $\delta^{13}\text{C} = -25\text{‰}$ .

*Siegenthaler*: Through simultaneous  $^{14}\text{C}$  and  $^{13}\text{C}$  measurements the relative contributions of fossil and of respiration  $\text{CO}_2$  could be estimated. How does this apply to your results on atmospheric  $\text{CO}_2$ ?

*Levin*: We calculated the respiration  $\text{CO}_2$  mixing ratio from a comparison of  $\Delta^{14}\text{C}$  and measured  $\text{CO}_2$  concentration (Levin, 1978). This led to admixtures about 3 times of the fossil fuel admixture in summer and

about the same as in winter time. Similar estimates could be made by simultaneous <sup>13</sup>C and <sup>14</sup>C measurements.

*Fritz:* Have you any information on <sup>14</sup>C variations with distance from a <sup>14</sup>C-emitting nuclear power station and have you compared <sup>14</sup>C with <sup>3</sup>H data?

*Levin:* 1) In tree-leaf samples from the environment of Würgassen and Obrigheim reactors we have measured <sup>14</sup>C increases only in the main wind direction and up to a distance of about 5km.

2) We have measured tritium in the washbottle water of the sampling system near the Biblis reactors and have not found unusual increases compared to the natural concentration.