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THE CONTINENTAL EUROPEAN SUESS EFFECT

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ABSTRACT. Observations of ^{14}C in atmospheric CO_2 at four different sites in central Europe, Heidelberg, Westerland, Schauinsland and Jungfraujoch have enabled us to determine individual fossil-fuel contributions to atmospheric CO_2 concentration. The data clearly show a decrease of fossil-fuel CO_2 with distance from anthropogenic source regions. At Heidelberg during winter we observe $^{14}C/^{12}C$ ratios up to 10% lower than at the clean air mountain station Jungfraujoch in the Swiss Alps, corresponding to an anthropogenic CO_2 contamination level of ca 10% at the Heidelberg site. The Schauinsland and Westerland winter fossil-fuel CO_2 concentrations are only ca 1.5 and 2% of the mean concentration, respectively. Our results indicate a strong seasonality in the European fossil-fuel CO_2 source with ca 50% lower CO_2 emissions during summer if compared to winter fossil-fuel CO_2 release. This effect may significantly contribute (by 1–2 ppm) to the observed annual cycle of atmospheric CO_2 concentration in northern mid-latitudes.

INTRODUCTION

In 1953 Hans Suess (1955; Revelle & Suess, 1957) discovered the so-called Suess Effect, which is a global decrease of $^{14}C/^{12}C$ ratio in atmospheric CO_2 found in plant material grown at the end of the 19th century. The reason for the "dilution" of ^{14}C in wood and respectively in atmospheric CO_2 is the input of fossil-fuel CO_2 into the atmosphere, which also has led to a remarkable increase of global atmospheric CO_2 concentration (Conway *et al,* 1988). The present effect of fossil-fuel CO_2 on the $^{14}C/^{12}C$ ratio in atmospheric CO_2 for the European continent, one of the most important sources of anthropogenic CO_2, is discussed here. The "regional" contributions observed at three sites in West Germany, a rural mountain station (Schauinsland/Black Forest), a coastal station (Westerland/Sylt) and a site in a highly populated area (Heidelberg) are compared to a source estimate of fossil-fuel CO_2 for this part of Europe. The results provide important new information for the validation of global carbon cycle models.

SAMPLING LOCATIONS

Heidelberg (49°N, 9°E). We collected continuous bi-weekly integrated CO_2 samples from ambient air at the Heidelberg Institute at 16m above ground. The Institute is on the outskirts of Heidelberg in the industrialized upper Rhine valley. A continuous record of $^{14}CO_2$ exists for the period of 1977–1988. Continuous measurements (1982–1983) of the radioactive noble gas ^{222}Rn are also available for the same sampling site. For the interpretation of the $^{14}CO_2$ results at Heidelberg, ^{222}Rn will be used as an atmospheric mixing tracer on the continental scale as well as for vertical mixing in the lower troposphere.

Schauinsland (48°N, 8°E). The WMO BAPMoN station Schauinsland run by the Federal Environmental Agency, UBA (Umweltbundesamt, Berlin), is close to the Schauinsland mountain top in the Black Forest. At 1200m asl, the station is usually above the ground level inversion layer of the nearby Rhine valley, surrounded by meadows and woods. In 1977 we started $^{14}CO_2$ sample collection and analyses of continuous bi-weekly samples taken from an inlet stack ca 7m above ground. Continuous measurements of CO_2 concentration, other atmospheric trace constituents and meteorological parameters are also available for the entire period of ^{14}C observations (UBA, 1982–1988). ^{222}Rn has also been continuously measured at the same site by the Institute of Atmospheric Radioactivity (Freiburg) for the period of $^{14}CO_2$ observations.

Westerland (55°N, 8°E). We started $^{14}CO_2$ measurements at the BAPMoN station Westerland in 1985. The station is situated at the North Sea island of Sylt, about 50m from the coast line in the dunes. Samples are collected from an inlet stack about 7m above local ground. Since this station belongs to the UBA network, the observation program is the same as at the Schauinsland station.

Jungfraujoch (47°N, 8°E). $^{14}CO_2$ collection started in 1986 at the High Alpine Research Station Jungfraujoch in the Swiss Alps. Because of its exposed position at 3500m asl, we mostly collect air that is not directly affected by the surrounding ground level sources (Zumbrunn *et al,* 1983). This effect occurs only in the summer when there is strong vertical mixing over the continents.

SAMPLING AND ANALYSIS

CO_2 samples were collected by quantitative absorption in sodium hydroxide solution. After transfer to the Heidelberg laboratory, CO_2 is extracted from the solution in a vacuum system by adding hydrochloric or sulfuric acid. For ^{14}C analysis, the CO_2 gas is purified over activated charcoal and counted in a high-precision proportional counter system. The $^{13}C/^{12}C$ ratio is measured by mass spectrometry from small aliquots of the CO_2 gas. Sampling and laboratory procedures are described in greater detail by Levin, Münnich and Weiss (1980) and Schoch *et al* (1980).

All ^{14}C activities are expressed as the per mil deviation ($\Delta^{14}C[‰]$) from the NBS oxalic acid activity corrected for decay (Stuiver & Polach, 1977). As all sampling and laboratory procedures were made nearly quantitatively, the $\Delta^{14}C$ values presented here are not corrected for fractionation of *individual* $\delta^{13}C$ values. To obtain a direct comparison with plant material, the values are corrected to $\delta^{13}C = -25‰$ using the mean value of all samples at one site for the observation period. The Schauinsland data were corrected, using a value of $\delta^{13}C = -8.13‰$, the Jungfraujoch data, $\delta^{13}C = -10.47‰$ (at this site the efficiency of the absorption was <100%), the Westerland data, $\delta^{13}C = -8.63‰$, and the Heidelberg data, $\delta^{13}C = -9.28‰$. The precision (1σ) of a single ^{14}C analysis is typically $\Delta^{14}C = \pm 4‰$.

DATA AND DISCUSSION

Figure 1 shows monthly mean ^{14}C concentrations calculated from bi-weekly data observed in atmospheric CO_2 from 1982 – 1988 at Jungfraujoch (A), Schauinsland (B), Westerland (C) and Heidelberg (D). For complete data from 1977 at Schauinsland and Heidelberg, see Levin (1987), Levin *et al,* (1988). A steady decrease (1987–1988: $\Delta^{14}C$ = -10.0‰ yr^{-1}) in $\Delta^{14}C$ was observed at all stations, which is partly due to the bomb ^{14}C spike still equilibrating with the ocean surface water and to the ongoing input of ^{14}C-

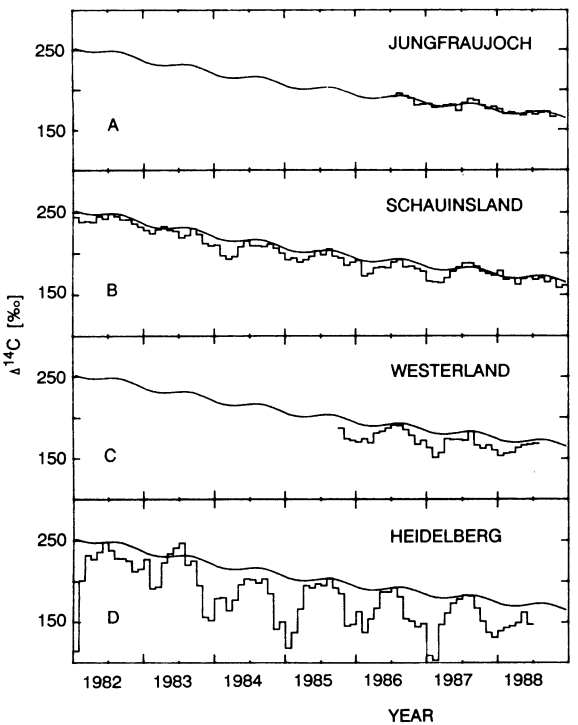

Fig 1. ^{14}C in atmospheric CO_2 observed at different sites in central Europe. Monthly mean values were calculated from continuous bi-weekly samples. The standard deviation of the monthly means is typically $\Delta^{14}C$ = 3‰ at stations Schauinsland, Westerland and Heidelberg, and $\Delta^{14}C$ = 2‰ at station Jungfraujoch. The smooth curve is the proposed 50°N $^{14}CO_2$ background level calculated from the observed Jungfraujoch level (1986–1988) and extrapolated backwards in time from the Schauinsland upper envelope with the Jungfraujoch seasonality of 1986–1988.

free fossil-fuel CO_2 into the atmosphere (the global Suess Effect). The individual stations, however, show significant differences in short-term fluctuations; at the Jungfraujoch, we observe a slight but significant seasonal cycle with lowest $^{14}C/^{12}C$ ratios during February and March and a maximum in August. The peak-to-peak difference is $\Delta^{14}C$ = 8‰. This seasonality may be partly due to a seasonally varying fossil-fuel CO_2 input (see discussion below). Part of the seasonality ($\Delta^{14}C$ = 2–3‰ (Tans, 1978)) may, however, be due to stratospheric intrusions of air enriched in ^{14}C into the troposphere, leading to a tropospheric ^{14}C maximum in July (compare with the early years after the bomb tests). Another source for a seasonal signal in tropospheric $^{14}CO_2$ is the biosphere, which, in recent years, generally acts as a net source of bomb $^{14}CO_2$ (Levin *et al,* 1987). The CO_2 flux from the biosphere into the atmosphere was measured by Dörr and Münnich (1987) at a site near Heidelberg. The CO_2 flux is highest in June/July (10mMol m^{-2} hr^{-1}) and

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lowest in February (1–2mMol m^{-2} hr^{-1}). The ^{14}C of this biogenic CO_2 shows a significant seasonality with a higher ^{14}C level in summer and ca 10% lower ^{14}C in the winter months (Dörr & Münnich, 1986). With a simple one-dimensional approach, we can estimate that the combination of the signal in CO_2 flux and $^{14}C/^{12}C$ ratio may lead to a total amplitude in atmospheric $^{14}CO_2$ of ca $\Delta^{14}C$ = 1–2‰ with a maximum in July/August.

We assume the ^{14}C level observed at the Jungfraujoch is representative of the undisturbed lower troposphere in mid-latitude westerlies (50°N) of the Northern Hemisphere. Deviation from this level at the other sites must then be attributed to an influence by "smaller scale" ground-level sources. Depending on the "clean air," respectively, "pollution" character of the Schauinsland, Westerland and Heidelberg sites, we mainly observe lower $\Delta^{14}C$ values and a more pronounced seasonal cycle than at the Jungfraujoch (Figs 1B, C, D). From this, namely that almost all deviations in $\Delta^{14}C$ at the "polluted" stations, if compared to the Jungfraujoch data, are in a negative direction, we interpret them to be due to "contamination" from continental European ^{14}C-free fossil-fuel sources – the "European Suess Effect." Hence, we assume that the biogenic as well as the nuclear power plant ^{14}C source (Levin *et al,* 1988) is of minor importance at these sites.

In order to quantitatively determine the seasonally varying contamination at the individual German sites, we have to define the representative ^{14}C background level at ca 50°N for the years before 1986 when no Jungfraujoch data exist. This has been done by extrapolating the Jungfraujoch record backwards in time. We used the mean decrease observed at Schauinsland and modulated this steady decrease with the smoothed seasonality observed at Jungfraujoch 1986–88 (smoothed curve in Fig 1A). In an earlier paper (Levin *et al*, 1985) we used the upper envelope of the Schauinsland record as the clean air background due to lack of data. Recent knowledge, particularly the results from this study (see below), however, show that the measured Jungfraujoch level should be a better approach to the $^{14}CO_2$ background at ca 50°N. Negative deviations from this "50°N background level" are now interpreted as due to regional fossil-fuel sources.

From Figure 1B, C, D, we conclude that the fossil-fuel effect during winter increases from ca 15‰ at the Schauinsland mountain site to ca 20‰ at the Westerland coastal site. At Heidelberg, where we are closest to ground level sources, the mean winter fossil-fuel contribution is ca 50‰ but can be up to 100‰ during ground-level inversions, often occurring during January and February in the Rhine valley. During the summer, the regional fossil-fuel contamination at all sites is considerably smaller, only ca 5‰ at Schauinsland and ca 10‰ at Westerland. The Heidelberg atmosphere is also found to be much less polluted in summer than in winter; the mean ^{14}C draw-down is only ca 10–15‰ from May to October. This effect is partly due to enhanced vertical mixing in the atmospheric boundary layer during summer and to a smaller fossil-fuel CO_2 input (see below).

Determination of the Fossil-Fuel CO_2 Concentration

The monthly mean fossil-fuel CO_2 concentration can be calculated from the observed $^{14}C_{obs}$ assuming a two component mixing of the observed atmospheric $CO_{2\,obs}$, namely $CO_{2\,obs} = CO_{2\,backgr} + CO_{2\,fossil}$:

$$CO_{2\,fossil} = CO_{2\,backgr} * (^{14}C_{backgr} - ^{14}C_{obs}) / ^{14}C_{obs} \quad (1)$$

with ^{14}C [pmc], CO_2 [ppm]: $^{14}C_{backgr}$ = extrapolated Jungfraujoch curve; $CO_{2\,backgr}$ = mean atmospheric CO_2 concentration at 50°N: 350 ppm (Conway *et al,* 1988).

Figure 2 shows the mean seasonal cycles of fossil-fuel concentrations at Schauinsland and Heidelberg for 1982–1988 and at Westerland for 1985 to 1988. The Schauinsland and Westerland records can be compared to monthly mean concentrations of atmospheric SO_2 measured at these sites parallel to $^{14}CO_2$. As a working hypothesis we assume that the SO_2 concentration at these sites predominently reflects anthropogenic emissions related to fossil-fuel burning (Cullis & Hirschler, 1980). Figure 3 shows the correlation between mean SO_2 concentration (UBA, 1982–1988) and fossil-fuel CO_2. At both sites there is a good correlation between SO_2 and fossil-fuel CO_2 derived from our ^{14}C measurements (correlation coefficient r = 0.85). This underscores the assumption that the ^{14}C variability observed at continental German stations is indeed due to fossil-fuel contamination. The observed SO_2 to CO_2 ratios at the individual stations Schauinsland (SO_2/CO_2 = 4.1μg m^{-3} ppm^{-1}) and Westerland (SO_2/CO_2 = 1.9μg m^{-3} ppm^{-1}) are, however, different. This may partly be due to the fact that SO_2, unlike CO_2, is not a conservative tracer of fossil fuel. The residence time of SO_2 in the atmosphere is highly variable, not exceeding several days. Thus, on its way from the sources to the observation site, SO_2 is partly oxidized to sulfate or dry-deposited. A better but still unconservative tracer for fossil fuel might be total sulfur concentration, mainly consisting of SO_2 and sulfate.

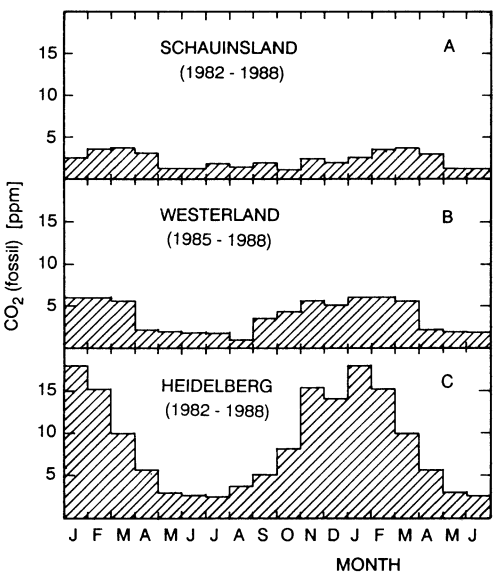

Fig 2. Monthly mean fossil-fuel CO_2 concentration at three German sites as calculated from the negative deviation of $^{14}CO_2$ from the proposed 50°N background (smooth curve in Fig 1). The influence from ground-level fossil-fuel sources decreases with the (horizontal or vertical) distance from the source regions: Heidelberg (C. Mean of 1982–1988); Westerland (B. Mean of 1985–1988); Schauinsland (A. Mean of 1982–1988).

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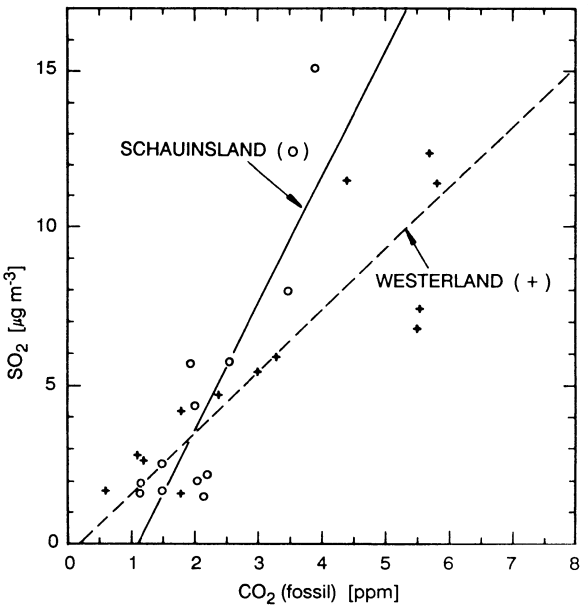

Fig 3. Correlation between monthly mean fossil-fuel CO_2 (Fig 2) and monthly mean SO_2 concentration at Schauinsland and Westerland. The correlation for both stations is quite good, but the slopes of the regression lines are different for the individual sites presumably due to SO_2 being partly oxidized on its way from the source regions to the observation sites.

Determination of the European Fossil-Fuel Source Strength

As stated previously, the difference of fossil-fuel CO_2 concentration at Heidelberg, if compared to clean air sites, eg, Schauinsland, is because Heidelberg is much closer to, or even within, ground-level anthropogenic sources. Particularly in winter, Heidelberg frequently lies in a strong ground-level inversion layer, whereas Schauinsland is much less affected by local ground-level sources of the Rhine valley (see, eg, Levin, 1987).

A natural tracer that can be used to parameterize the "contamination" of an air mass by continental ground-level sources is the soil-borne radioactive noble gas ^{222}Rn ($T_{1/2}$ = 3.8 days) (Dörr *et al,* 1983; Volpp, 1984). ^{222}Rn is produced in soils, the emanation rate of radon from the surface ocean is of only minor importance. The only sink of ^{222}Rn is radioactive decay. The emanation rate of ^{222}Rn from European soils is constant with time and does not show spatial variability larger than a factor of two (Dörr, 1984). Thus, with the mean radon emanation rate, measurements of the atmospheric ^{222}Rn activity concentration, in principle, lead to a measure of the so-called atmospheric mixing height, respectively, the travelling time of an air mass over the continent (see, eg, Levin, 1987).

The monthly mean ^{222}Rn activity observed at Schauinsland station from 1982–1988, and at Heidelberg from 1982–1983 is plotted in Figure 4. For the further calculations, the Heidelberg record is extended for the years 1984–1988 using measurements from Freiburg, a site which is also in the Rhine valley ca 170km south of Heidelberg. The data from Schauinsland and Freiburg were provided by the Institute of Atmospheric Radioactivity, Freiburg (Sartorius, pers commun); for the Heidelberg radon activities see Volpp (1984). Figure 4 shows that the variability of the Heidelberg (solid

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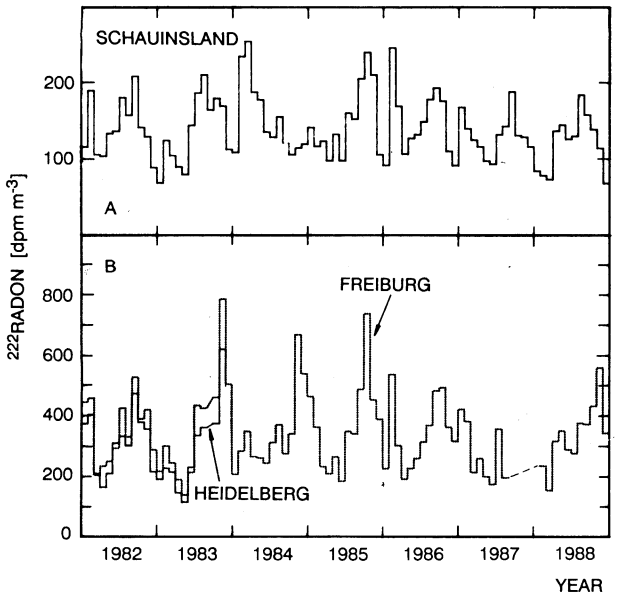

Fig. 4. Monthly mean ^{222}Rn activities at Schauinsland (A. 1982–1988 (Sartorius, pers commun)), Heidelberg (B. 1982–1983, solid line (Volpp, 1984)) and Freiburg (B. 1982–1988, dotted line (Sartorius, pers commun)). Correlation of the 1982–1983 monthly mean atmospheric ^{222}Rn activities at Heidelberg and at Freiburg leads to a slope of the regression line Freiburg/Heidelberg = 1.19; the correlation coefficient is r = 0.97.

line) and Freiburg (dotted line) ^{222}Rn levels compares very well; the correlation of the monthly mean values at the two Rhine valley sites for 1982–1983 leads to a correlation coefficient r = 0.97, the Freiburg level ca 19% higher than the Heidelberg level. The latter effect may be due to a slightly different emanation rate at Freiburg if compared to Heidelberg. Thus for the following calculations, we used the Freiburg monthly mean ^{222}Rn values divided by a factor of 1.19. The absolute difference of the ^{222}Rn activity between the Rhine valley sites, Heidelberg/Freiburg, and the mountain site, Schauinsland, is evident and nicely illustrates the vertical radon gradient. The strong seasonality, particularly at the Rhine valley sites, with high ^{222}Rn activity during autumn and winter, reflects the strong night and winter-time inversions in the Rhine valley during this part of the year (Volpp, 1984).

Assuming the ^{222}Rn emanation rate j_{Rn} to be homogeneous over the European continent and constant with time, we can calculate the fossil-fuel CO_2 flux with a very simple one-dimensional atmospheric model. For this, we use the observed ^{222}Rn activities [Rn] at Schauinsland and Heidelberg/Freiburg, and the ^{14}C-derived fossil-fuel CO_2 concentration $[CO_{2\,fossil}]$:

$$j_{fossil} / j_{Rn} = [CO_{2\,fossil}] / [Rn]. \quad (2)$$

With this approach we, implicitly, assume that both sources, j_{Rn} and j_{fossil}, are similarly homogeneously distributed on the continent, which, of course, for the fossil-fuel source, is only partly true on a large scale.

The mean ^{222}Rn emanation rate from European soils is $j_{Rn} = 4000 \pm 2000$ dpm m^{-2} hr^{-1} (Dörr & Münnich, 1989). The emanation rate from the sandy soils in the Rhine valley, particularly around Heidelberg, is, however, at the lower end of that range, amounting to 2500 ± 1000 dpm m^{-2} hr^{-1} (Dörr & Münnich, 1987). Thus, we use the latter number for the calculation of the

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Heidelberg fossil-fuel source. For Schauinsland, however, with its considerably larger fetch, *ie,* all of western Europe, we use the mean European ^{222}Rn flux of 4000 dpm m^{-2} hr^{-1}. The result of our calculations of j_{fossil} is plotted in Figure 5.

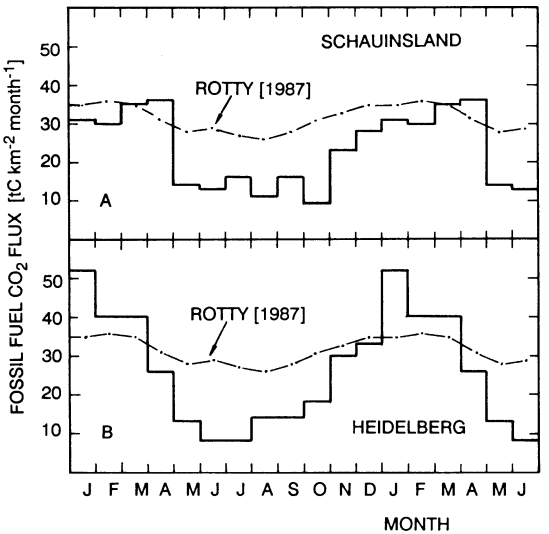

Fig 5. Seasonal cycle of the fossil-fuel source strength as calculated from the fossil-fuel contribution at Schauinsland (A) and Heidelberg (B) (Fig 2) and the radon activity concentrations at the same sites (Fig 4) according to equation (2). Also given is the mean seasonality reported by Rotty (1987) (smooth dotted line).

The yearly mean fossil-fuel source strength in the environment of Heidelberg and Schauinsland turn out to be 296 tC km^{-2} a^{-1} and 262 tC km^{-2} a^{-1}, respectively. These numbers are consistent with each other but slightly lower than Rotty's (1987) estimate (373 tC km^{-2} a^{-1}) if we assume that Heidelberg and Schauinsland are mainly influenced by westerly winds, and, thus, mainly see the mean fossil-fuel source of West Germany and France. The reason for the difference in absolute source strength to Rotty's number, if significant, may be that, using the Jungfraujoch site as a reference in the above calculations, we obtain only the *additional* (continental European) fossil-fuel contamination for Schauinsland and Heidelberg, which, of course, has to be smaller than the *total* source in this latitude belt.

Another important feature of the above estimate is that we find a much more pronounced seasonality in the fossil-fuel CO_2 release than Rotty (1987) estimates; which may be because he only uses the fossil-fuel sales statistics instead of real consumption statistics. Since the energy demand for private homes, particularly in Europe, has a strong seasonality and is ca 40% of the total energy consumption (Bartholomäi & Kinzelbach, 1980), the observed seasonality is easily explained. We should also keep in mind that the seasonality and the mean value of the fossil-fuel CO_2 concentration, particularly at Schauinsland, strongly depend on the determination of the ^{14}C reference level for which we chose the Jungfraujoch. There is no doubt that even the Jungfraujoch is, to a certain extent, influenced by ground-level anthropogenic sources. This influence should be *largest* during the summer (see, *eg,* Dams & de Jonge, 1980) when vertical mixing over the

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continent is enhanced. However, we do observe at the Jungtraujoch *lowest* ^{14}C levels during the winter (*cf* Fig 1A) which is another strong argument for the Jungfraujoch as a useful reference site for northern mid-latitudes.

CONCLUSIONS

High-precision ^{14}C measurements on atmospheric CO_2 are a powerful tool for determining the influence of fossil-fuel CO_2 on the CO_2 budget over highly industrialized continental regions. From this study, we conclude that the mean source strength of fossil-fuel CO_2 necessary *in the yearly mean* to explain the observed fossil-fuel CO_2 concentrations compares quite well with the direct estimate. However, the *seasonality* of the fossil-fuel input is considerably larger in the European source region than has been estimated from the data of Rotty (1987). The seasonally varying input may, thus, have a considerable effect on the amplitude of the Northern Hemisphere seasonal CO_2 concentration cycle. If ca 2/3 of the total Northern Hemisphere fossil-fuel source, namely the total release from Europe, USSR and China, would have a relative seasonality by a factor of two as concluded from our findings, this could lead to an absolute seasonality of 1–2 ppm in 50°N CO_2 concentration. The observed increase of the amplitude of the seasonal cycle of atmospheric CO_2 in the Northern Hemisphere in the last 25 years (Conway *et al,* 1987), therefore, may, to some extent, be due to the increasing fossil-fuel CO_2 source. Moreover, the smooth seasonality of $^{14}CO_2$ at the Jungfraujoch station of $\Delta^{14}C$ = 8‰ can, to a large extent (ca 50%), be explained by the seasonality of the Northern Hemisphere fossil-fuel source.

High-precision ^{14}C measurements of atmospheric CO_2 open an independent means to deconvolve the seasonal cycle of atmospheric CO_2 in the Northern Hemisphere because they provide a unique possibility to distinguish between recent biogenic and anthropogenic sources which are both very important for the Northern Hemisphere atmospheric CO_2 budget.

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