The Schauinsland CO_2 record: 30 years of continental observations and their implications for the variability of the European CO_2 budget

M. Schmidt,[1,2] R. Graul,[3] H. Sartorius,[4] and I. Levin[1]

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[1] Since 1972, the German Environment Agency (UBA) has been measuring continuously CO_2 concentration at Schauinsland station (southwest Germany, 1205 m asl). Because of its vicinity to biogenic and anthropogenic sources and sinks, the Schauinsland CO_2 record shows considerable variability. In order to remove these disturbances and derive the large-scale representative ''background'' CO_2 level for the respective area (southwest Germany) we perform rigorous data selection based on wind speed and time of day. During the past 30 years, the selected CO_2 mixing ratios increased by 1.47 ppm per year, following the mean trend in midlatitudes of the Northern Hemisphere. The average seasonal cycle (peak to peak) amplitude has decreased slightly from 13.8 ± 0.6 ppm in the first decade (1972–1981) to 12.8 ± 0.7 ppm in the last two decades (1982–2001). This is opposite to other northern latitude sites and is attributed to the decrease of fossil fuel CO_2 emissions in the catchment area (southwest Germany and France) and its respective change in the seasonal variation. Except for May and June, monthly mean CO_2 mixing ratios at Schauinsland are higher by up to 8 ppm if compared to marine boundary layer air, mainly as a consequence of fossil fuel CO_2 emissions in Europe. The CO_2 measurements when combined with continuous ^{222}Rn observations at the same site allow an estimate of the net CO_2 flux in the catchment area of Schauinsland: Mean seasonal fluxes compare very well with estimates from a process-oriented biosphere model (SIB-2) as well as from an inverse modeling approach [*Peylin et al.*, 2000]. Annual CO_2 fluxes vary by more than a factor of 2, although anthropogenic fossil fuel CO_2 emissions show interannual variations of only about 10%. The major part of the variability must therefore be associated to interannual changes of biospheric uptake and release, which are on the order of the total fossil fuel emissions in the same area. This has to be taken into account when reliably quantifying and verifying the long-term carbon balance and emission reduction targets in the European Union. *INDEX TERMS:* 0315 Atmospheric Composition and Structure: Biosphere/atmosphere interactions; 1610 Global Change: Atmosphere (0315, 0325); 9335 Information Related to Geographic Region: Europe; *KEYWORDS:* carbon dioxide, radon, europe, CO_2 fluxes, biosphere.

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1. Introduction

[2] Over the past three decades the annual accumulation of CO_2 in the atmosphere has varied between 1 and 6 PgC per year [*Conway et al.*, 1994]. The year-to-year variability of fossil fuel emissions has, however, been small (<0.3 PgC yr^{-1} [*Marland et al.*, 2002]); therefore the observed changes in atmospheric carbon accumulation mainly reflect variations of ocean and land fluxes. Several atmospheric CO_2 measurement networks have been established since the pioneering work by Keeling in 1957/58 at Mauna Loa, Hawaii, and at the South Pole [*Keeling*, 1993]. At present, about 120 data records from all over the globe are available [*GLOBALVIEW-CO_2*, 2002]. Most of the observational stations do, however, sample marine boundary layer air, and only a limited number of sites are located on the continent, and are able to directly capture continental biospheric and fossil fuel CO_2 emissions signals.

[3] The Global Atmosphere Watch (GAW) station Schauinsland in the Black Forest in southwest Germany is one of these continental sites with a more than 30 yearlong history of atmospheric CO_2 observations [*Levin*, 1987]. The station has the longest continental CO_2 record available to the scientific community and is thus ideal to study long-term and interannual variations of CO_2 sources and sinks in

[1]Institut für Umweltphysik, Universität Heidelberg, Heidelberg, Germany.
[2]Now at Laboratoire des Sciences du Climat et l'Environnement, CE Saclay, Gif-sur-Yvette, France.
[3]Umweltbundesamt, Messstelle Schauinsland, Kirchzarten, Germany.
[4]Bundesamt für Strahlenschutz, IAR, Freiburg, Germany.

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Western Europe. However, due to the close vicinity of this site (and generally all ground-based continental stations) to those continental sources and sinks we are interested in, it is difficult to make measurements here which are representative of large spatial scales. Aircraft would probably have been a more adequate platform for such observations over continents [*Tans et al.*, 1996], however, high frequency aircraft sampling is extremely expensive and does not provide continuous records. Alternatively, observations on very tall towers have been started in the last decade [*Bakwin et al.*, 1995, 1998]. Still, the classical sites in the continental realm are mountain stations, despite the difficulties to characterize their meteorological regime and compare their observations with model estimates [e.g., *Chevillard et al.*, 2002a, 2002b]. In the present study we try to overcome these inherent problems of continental mountain sites by introducing a rigorous data selection procedure to eventually derive a large-scale representative atmospheric CO_2 record.

[4] In addition to the continuous CO_2 data we use concurrent ^{222}Rn observations to estimate CO_2 net fluxes by applying the ^{222}Rn tracer method [*Levin*, 1984; *Schmidt et al.*, 1996; *Wilson et al.*, 1997; *Levin et al.*, 1999; *Biraud et al.*, 2000; *Schmidt et al.*, 2001]. ^{222}Rn is a radioactive noble gas with a lifetime of 5.5 days, which is produced naturally at relatively constant rates in all soils and is diffusing to the atmosphere, where it is diluted by transport and radioactive decay. The ^{222}Rn flux from ocean surfaces is almost negligible. The atmospheric ^{222}Rn activity can thus be used to parameterize continental air mass residence times and provides a quantitative measure of the dilution of continental ground level emissions in the atmospheric surface layer driven by atmospheric mixing. The ^{222}Rn tracer method is applied here to selected CO_2 and ^{222}Rn data, excluding influences from local sources and sinks, to estimate mean continental CO_2 fluxes representative for southwest Germany and southern France.

2. Methods

2.1. Sampling Site

[5] The continental GAW station Schauinsland (47°55′N, 7°55′E, 1205 m asl) is part of the atmospheric monitoring network of the German Environment Agency, Berlin (UBA). The station is situated on a mountain ridge in the Black Forest, in southwest Germany, at an elevation of about 1000 m above the highly populated Rhine valley. During night, the station is usually sampling air from above the Rhine valley inversion layer, while during the day, particularly in summer, the Schauinsland station frequently measures air masses influenced by local CO_2 sources and sinks. The station is surrounded by meadows and woods; during winter the ground is largely snow covered.

2.2. CO_2 Analysis

[6] Atmospheric CO_2 concentration has been continuously monitored at Schauinsland since 1972 by nondispersive infrared analysis (NDIR) [*Levin*, 1987]. Until August 1980, CO_2 measurements were performed with URAS-2 (Hartmann & Braun), from September 1980 until the end of 1993 with Ultramat-3 (Siemens) and from 1994 onward with URAS-3 (Hartmann & Braun). The data obtained until 1991 were calibrated with CO_2-in-N_2 standard gases and are therefore corrected for carrier gas effect [*Levin et al.*, 1995]. Later measurements were performed with CO_2-in-air standard gas mixtures. All CO_2 data are reported on the WMO CO_2 mole fraction scale. The accuracy of the CO_2 concentration data is estimated to better than ±1 ppm for the period 1972–1991 and better than ±0.5 ppm later on. In an attempt to assess differences in standard scales among laboratories contributing CO_2 data to global data bases and to *GLOBALVIEW-CO_2* [2002], WMO has initiated two laboratory intercomparison experiments. In the 1995/1996 and 1998/1999 experiments CO_2 measurements at Schauinsland laboratory agreed to better than 0.15 ppm with the WMO Central CO_2 Calibration Laboratory [*Peterson et al.*, 1999].

2.3. Radon 222 Analysis

[7] Radon 222 is measured at the Schauinsland site by the Bundesamt für Strahlenschutz at a station ca. 150m away from the UBA station. Radon is measured with the so-called static filter method [*Stockburger and Sittkus*, 1966]: ambient air is continuously pumped through a quartz fiber filter, where the ^{222}Rn daughters that are attached to aerosol particles are quantitatively retained. The α decay of the ^{222}Rn daughters ^{218}Po and ^{214}Po is counted in situ, and the net atmospheric ^{214}Po activity concentration is then calculated from the ^{214}Po activity on the filter. A mean disequilibrium (mean ratio between ^{214}Po and ^{222}Rn activity) of 0.85 ± 0.14 was estimated for this measurement site [*Schmidt*, 1999]. This value is used here to calculate the atmospheric ^{222}Rn activity from the observed atmospheric ^{214}Po activity.

2.4. Data Selection

[8] In order to remove locally influenced data from those representative for a larger area over the European continent, we perform a selection procedure on the Schauinsland CO_2 and ^{222}Rn measurements which was previously described by *Schmidt et al.* [1996]. To avoid source/sink influence from the Rhine valley through upslope winds during the day, we use only nighttime values (2200–0600 LT) and select situations with high wind speed (>2.5 m sec^{-1} in summer and >3.5 m sec^{-1} in winter). The calculated daily mean CO_2 mixing ratio is finally accepted if more than 8 half hourly values have been selected and the CO_2 standard deviation of these 8 or more values is smaller than 1.5 ppm. On average 87 ± 21 days per year satisfy this selection criterion between 1972 and 2001. The selected data are distributed almost evenly between all months with July and August data being slightly underrepresented (July/August 5.5%, all other month 7–10%). We employ a curve fitting procedure to the selected data described by *Thoning et al.* [1989] and *Masarie and Tans* [1995]. The curve fit incorporates harmonic and quadratic functions, and an 80-day smoothing is applied to the residuals. A 3-sigma filter is applied in the smoothing process to obtain the best-fit curve, excluding statistical outliers.

3. Results and Discussion

3.1. CO_2 Observations

[9] Figure 1a shows selected daily mean CO_2 mixing ratios for Schauinsland station from 1972 to 2001 together with the fitted curve through the selected data. Crosses are

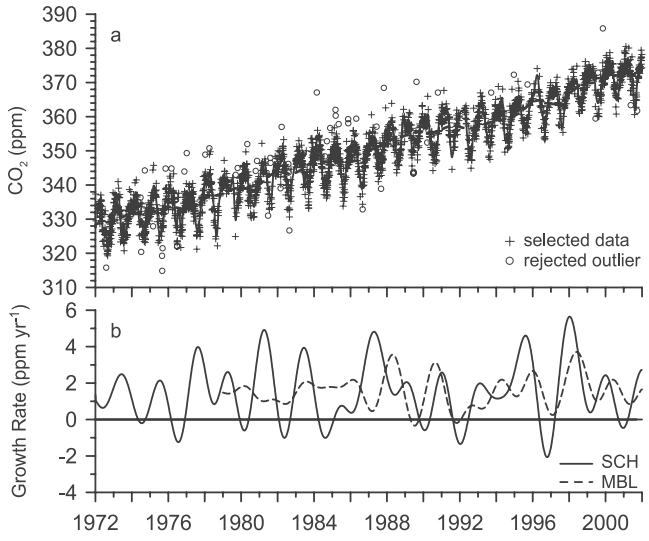

Figure 1. (a) Selected daily mean CO_2 data for Schauinsland station. The solid curve results from an iterative fit to the data excluding values, which deviate by more than three standard deviations from the fit. Crosses are those data used in the final fit, circles were statistically excluded. The dashed line shows the deseasonalized trend curve. (b) Annual growth rates as derived from the deseasonalized trend curve at Schauinsland (solid line) in comparison to the respective growth rate observed in the marine boundary layer at 48°N (dashed line is from *GLOBALVIEW-CO_2* [2002]).

data used in the final fit; circles represent data statistically excluded from the fit as outliers. The annual mean CO_2 mixing ratio increased from 328.3 ppm in 1972 to 372.3 ppm in 2001. This results in a mean annual increase rate of 1.47 ppm for the last 30 years. The dashed curve in Figure 1a represents the trend curve with the seasonal cycles removed. Still, there is significant interannual variation apparent in the CO_2 growth rate, which is shown in Figure 1b. Throughout the last 30 years significant negative growth of less than -1 ppm yr^{-1} is observed in 1976, 1991/1992 and 1996, and higher than average growth during 1980/1981, 1987 and 1997/1998. After a general decline in the early 1990s extreme variations are found in the growth rate from 1995 onward. The growth rate anomalies observed between 1995 and 2001 at Schauinsland station are also found in the marine boundary layer of the latitude belt of 48°N (Figure 1b, dashed line [*GLOBALVIEW-CO_2*, 2002]). The amplitudes of the global anomalies, however, are about 1.5 times smaller in 95/96 and 1998 and are small but remain positive in 1997 and 1999 [*Tans et al.*, 1998] (see http://www.cmdl.noaa.gov/ccgg). The slightly negative growth rates frequently observed at Schauinsland in the first half of the 1980s are not found in the marine boundary layer reference. They may be due to regional (European) events or not present in the marine reference, because this record is biased by Pacific sites during the 1980s.

[10] Regular seasonal variations, i.e., low CO_2 mixing ratio during summer and high mixing ratio in winter are well developed. On average over the observation period, we find a mean amplitude (peak-to-peak) of 13.2 ± 1.2 ppm. The seasonal amplitude seems to be slightly decreasing during the period of observations from 13.8 ± 0.6 ppm in the first decade (1972–1981) to 12.8 ± 0.7 ppm in the last two decades (1982–2001). The decrease of the CO_2 amplitude in the last two decades is opposite to findings at other northern latitude stations [*Keeling et al.*, 1996; *Myneni et al.*, 1997] but, if significant, would be consistent with a decrease of the fossil fuel CO_2 contribution and its seasonal amplitude at Schauinsland station by ca. 20% as derived from $^{14}CO_2$ observations [*Levin and Kromer*, 1997; I. Levin et al., A novel approach for independent budgeting of fossil fuel CO_2 over Europe by $^{14}CO_2$ observations, submitted to *Geophysical Research Letters*, 2003].

3.2. Comparison of the Schauinsland Data With the Marine Reference

[11] The CO_2 signal at Schauinsland station contains the hemispheric CO_2 background variability and trend but also influence from large-scale biospheric sources and sinks as well as from fossil fuel emissions. In order to quantify this continental source/sink influence, we compare our CO_2 record to the marine boundary layer (MBL) reference curve for 48°N which is derived from the global CO_2 measurement network [*GLOBALVIEW-CO_2*, 2002]. Figure 2a shows the selected and smoothed curve derived from Schauinsland data (solid line) in comparison to MBL (dashed line) for the period 1980 to 2001. The general shape of both records is similar, however, a closer look exhibits a small phase shift, which is visible when examining the monthly differences between the curves plotted in Figure 2b. These differences are most probably a consequence of the continental biospheric sources and sinks.

[12] In most years we observe a CO_2 mixing ratio at Schauinsland being higher than the marine reference during autumn and winter, and lower in spring to summer (May and June). The mean seasonal cycle of the CO_2 offset is shown in Figure 3a. During spring and summer, air masses coming from the Atlantic Ocean and traveling over the European continent get depleted in CO_2 due to the assimilation sink. The negative offset indicates that the catchment area of Schauinsland station in spring and early summer represents a net CO_2 sink. During the rest of the year, the CO_2 offset at Schauinsland is mainly positive, indicating that the fossil fuel and biospheric sources are stronger than the assimilation sink. The seasonal amplitude of the CO_2 offsets, however, varies considerably from year to year; the same is true for the annual mean concentration offset which varies between 0.51 and 1.38 ppm in the period of observations. These differences can be due to interannual variations in sources and sinks but also due to year to year changes in residence time of the air masses over the continent arriving at Schauinsland. Trajectory analyses for the selected data show that 95% of the air masses reach the Schauinsland station from western to southwestern direction and only 5% from the north or east. The catchment area of the Schauinsland station during well-mixed situations is thus assumed as southwest Germany and France.

3.3. Radon 222 as a Tracer to Estimate Air Mass Residence Times

[13] In order to estimate transport processes and residence times of the air masses over the source and sink areas, and for deriving CO_2 flux densities for the catchment area of the

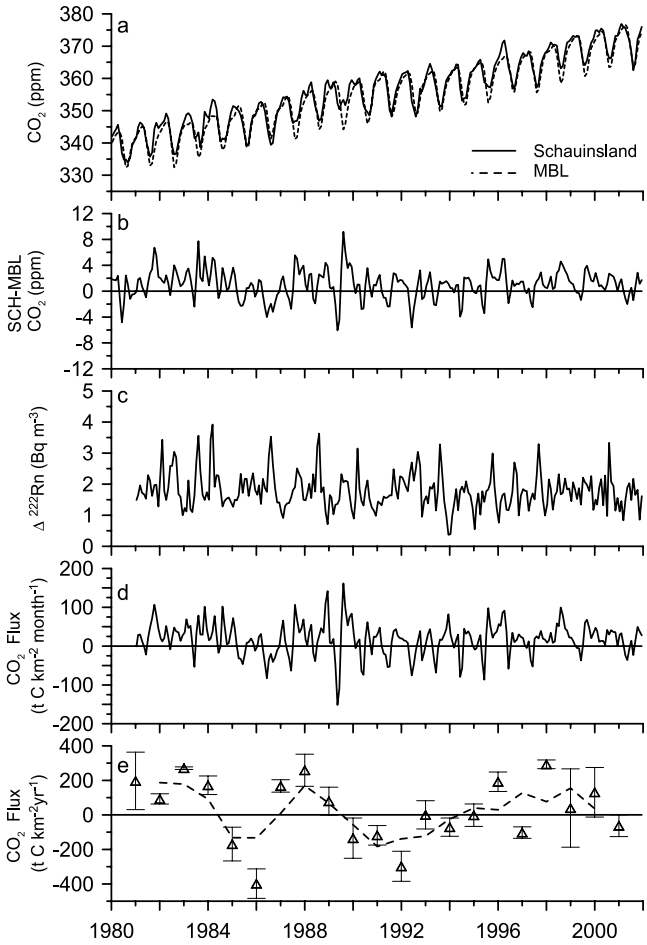

Figure 2. (a) Selected and smoothed CO_2 record from Schauinsland station (solid line) in comparison to the marine reference curve (MBL, dashed curve) for 48°N [*GLOBALVIEW-CO$_2$*, 2002]. (b) Concentration difference between the smoothed Schauinsland and the MBL curve. (c) The ^{222}Rn gradient between marine air (set constant at 0.3 Bq m^{-3}) and selected monthly data observed at Schauinsland. (d) Total net CO_2 flux calculated with the ^{222}Rn tracer method for the catchment area of the Schauinsland site. (e) Annual mean biogenic CO_2 flux in the catchment area of Schauinsland; the dashed line represents a three-point running mean.

Schauinsland site, we used radon 222 as tracer. Monthly mean ^{222}Rn activity concentrations vary between 0.5 and 4 Bq m^{-3} for the periods where CO_2 data were selected (Figure 2c). During December and January ^{222}Rn activity is generally lowest indicating decoupling of air masses from ground level sources. From July to September ^{222}Rn exhibits higher concentration values than average corresponding to on average longer continental residence times and/or stronger vertical mixing of air masses over the European continent, transporting more ^{222}Rn to the Schauinsland mountain site (Figure 3b).

[14] For our CO_2 source estimates we apply the continental approach of the ^{222}Rn tracer method as described by *Schmidt et al.* [2001], which focuses on atmospheric concentration changes during the large-scale transport of air masses over the European continent. We subtract the marine reference concentration from the selected Schauinsland CO_2 data, and, in an equivalent way, we treat the selected ^{222}Rn data (where ^{222}Rn in marine air is assumed to be constant at 0.3 Bq m^{-3} [*Biraud et al.*, 2000]). We then assume that the respective offsets reflect the sources and sinks of the two gases along the path of the (marine) air mass moving across the European continent. *Schmidt et al.* [2001] derived the simplified equation for the CO_2 flux estimate with the ^{222}Rn tracer method to

$$\bar{j}_{CO2} = \bar{j}_{Rn} \frac{\Delta c_{CO2}}{\Delta cRn} \cdot \left(1 - \frac{\lambda Rn \cdot cRn}{\frac{\Delta cRn}{\Delta t}}\right) \quad (1)$$

with $\bar{j}_i$ being the respective trace gas flux densities, Δc_i the monthly differences between marine reference and Schauinsland observations (Figures 2b and 2c), λ_{Rn} the decay constant for ^{222}Rn and Δt the traveling time of the air mass from the ocean to the measurement site.

3.4. Estimate of the Mean ^{222}Rn Exhalation Rate and Decay Correction for the Schauinsland Catchment Area

[15] The ^{222}Rn exhalation rate j_{Rn} from continental soil surfaces strongly depends on the texture of the soils, but studies show quite homogeneous and temporarily constant behavior in a restricted area [*Dörr and Münnich*, 1990]. Higher soil humidity leads to slightly diminished exhalation rates in fine-grained soils during winter [*Schüssler*, 1996]. From the distribution of the soil texture [*Food and Agricultural Organization* (*FAO*), 1981] in our assumed catchment area, i.e., France, and a large number of flux measurements in different soil types [*Schüssler*, 1996; *Jutzi*, 2001] we calculate a yearly mean exhalation rate of 61.2 Bq m^{-2} h^{-1}. A seasonal variation with 20% higher emissions in summer and 20% lower emissions in winter was derived from long-term flux measurements at five sites close to Heidelberg with different soil texture [*Dörr and Münnich*, 1990; *Schüssler*, 1996; *Jutzi*, 2001].

[16] Trajectory analyses yield a mean residence time of 3 ± 1 days for air masses over the European continent

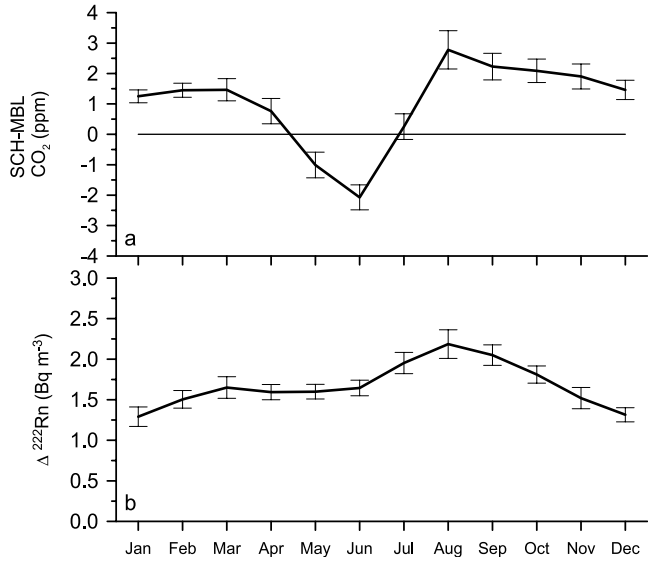

Figure 3. (a) Mean seasonal cycle of the CO_2 offset between Schauinsland and MBL (derived from Figure 2b). (b) Mean seasonal cycle of the corresponding ^{222}Rn offset (derived from Figure 2c).

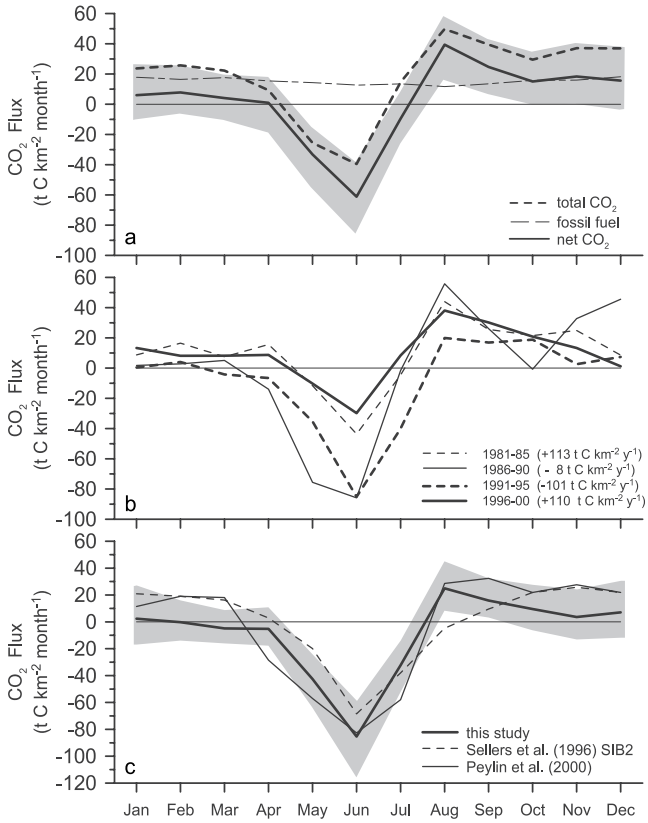

Figure 4. (a) Mean seasonal cycle of the total radon-derived CO_2 flux (solid line) together with the mean fossil fuel flux (dotted-dashed line) [*Marland et al.*, 2002; *Rotty*, 1987] and the difference between both curves interpreted as biogenic CO_2 flux. The shaded area corresponds to the uncertainty range (1 sigma) of the biogenic CO_2 flux estimate. (b) Mean seasonal biogenic CO_2 fluxes derived for four 5-year periods. Annual mean CO_2 fluxes for the four periods are reported in the legend. (c) Mean seasonal biospheric CO_2 flux in the catchment area of Schauinsland for the years 1990–1995 in comparison to the biosphere model SiB2 [*Sellers et al.*, 1996] and an inverse modeling approach by *Peylin et al.* [2000] for Europe. The shaded area corresponds to the uncertainty range (1 sigma) of the radon derived CO_2 flux estimates for the period of 1990–1995.

before reaching Schauinsland station. For a continental residence time of 2–4 days the net effect of radioactive decay (term in brackets in equation (1)) then corresponds to a loss of 16–29%. As the variation of this correction is on the order of 10% or less, we applied a constant decay correction factor of 0.77.

3.5. CO_2 Flux Estimates

[17] The ^{222}Rn tracer method has been applied to the monthly CO_2 and ^{222}Rn offsets between Schauinsland and MBL, and a CO_2 flux was estimated for the catchment area (Figure 2d). Negative CO_2 concentration offsets translate into negative CO_2 fluxes (uptake by plant assimilation) during spring and early summer. Because of the large variability observed for the monthly concentration offsets, the monthly flux estimates show a large variability up to a factor of two in the seasonal amplitudes but also in the annual mean values. The mean seasonal cycle for the whole period of observations is presented in Figure 4a. This mean flux estimate consists of biospheric emissions and uptake (respiration and assimilation) but also of fossil fuel CO_2 emissions. In May and June assimilation dominates the net CO_2 flux and results in a long-term mean net monthly CO_2 uptake of about 40 tC km^{-2} month^{-1} (40 10^6 gC km^{-2} month^{-1}).

[18] Elimination of the fossil fuel CO_2 contribution from the record can in principle, be performed using our $^{14}CO_2$ measurements [*Levin et al.*, 1989; *Levin and Kromer*, 1997]. However, these measurements provide only values integrated over 14 days and they can thus not be selected for background situations as is the case for the CO_2 data. Therefore we apply a different approach, and use the fossil fuel CO_2 emissions from a statistical data base [*Marland et al.*, 2002] to derive only the biogenic contribution to the total CO_2 flux. As mentioned earlier, France most likely reflects the catchment area of the selected Schauinsland CO_2 record. We therefore subtracted the fossil fuel CO_2 emissions of France to estimate the biospheric net CO_2 flux in the catchment area of Schauinsland.

[19] *Marland et al.* [2002] compiled fossil fuel CO_2 emissions from individual countries on an annual basis. Fossil fuel emissions do, however, also exhibit a seasonal variation, which was estimated by *Rotty* [1987]. Taking this seasonality into account, all monthly mean radon-derived flux data were corrected for fossil fuel contribution, then allowing for an estimate of the solely biogenic flux. The mean seasonal variation of this biogenic flux for the period of 1980–2001 is also displayed in Figure 4a together with the mean seasonal fossil fuel flux. It is interesting to note that even during the winter months (January and February), biogenic fluxes contribute about 30% to the total CO_2 flux. This finding is in agreement with results of *Levin et al.* [1980] derived from $^{14}CO_2$ observations.

[20] As already observed for the total flux, the purely biogenic net CO_2 flux shows considerable variability from year to year, while the fossil fuel emissions are decreasing from 1981 (195 tC km^{-2} year^{-1}) to 1985 (169 tC km^{-2} year^{-1}), showing only a small variation (155–173 tC km^{-2} year^{-1}) thereafter. As illustrated in Figure 2e annual fluxes are negative in 1985 and 1986 as well as in 1990 to 1992. The latter period was characterized also by an anomalously low CO_2 growth rate in marine boundary layer air (see Figure 1b). For a further inspection of the variability of the biogenic net flux, CO_2 fluxes have been grouped into four different five year periods and the mean seasonal cycles are plotted in Figure 4b. While autumn and winter fluxes are rather stable, largest variability between the five-year groups is observed during spring and summer when gross exchange fluxes between the atmosphere and the biosphere are largest. At this time of the year the variability is on the order of the total net flux (40–80 tC km^{-2} month^{-1}), and leads to variations in the Schauinsland catchment area of the mean annual net biogenic flux averaged over five years changing from a small net source of 113 tC km^{-2} year^{-1} (1981–1985) to a small net sink of −101 tC km^{-2} year^{-1} (1991–1995).

3.6. Estimate of the Uncertainty of the Radon-Derived CO_2 Fluxes

[21] The uncertainty of the radon-derived CO_2 flux estimates is mainly determined by the uncertainty of (1) the

marine boundary layer reference curve, (2) the intercomparability of the Schauinsland data with the GLOBALVIEW-CO_2 data set, and (3) the uncertainty of the ^{222}Rn flux and ^{222}Rn activity determination.

[22] 1. The number of stations included in the calculation of the MBL changes during the course of our observations. During the 1980s the MBL is biased toward Pacific sites while in the 1990s Atlantic and Pacific sites contribute almost equally to the MBL reference. To investigate the effect of Pacific versus Atlantic sites we calculated an Atlantic MBL curve for the periods 1981–1985 and 1996–2000 solely from data of the stations Azores and Station P and compare it to the standard MBL determined from all sites. Monthly mean differences between the two curves were well below 1 ppm, the standard deviation of the difference smaller than 0.5 ppm.

[23] 2. Intercomparability of Schauinsland observations with the GLOBALVIEW-CO_2 data set is estimated to about 0.5 ppm for the first half of the observational period (1980–1990) where systematic scale differences could be caused by errors in the carrier gas correction. For the second half of the observational period intercomparability was much better, on the order of ±0.2 ppm as documented by the performance of the Schauinsland laboratory in the WMO Round Robin Intercomparison exercises [*Peterson et al.*, 1999].

[24] 3. The uncertainty of the atmospheric ^{222}Rn activity observations is smaller than ±15%. This includes the uncertainty of the atmospheric disequilibrium factor applied to derive the ^{222}Rn activity from radon daughter measurements. The uncertainty of the absolute calibration of the atmospheric ^{222}Rn measurements is not contributing to the overall error of the CO_2 flux determination because the ^{222}Rn flux measurements from soils are on the same scale as the atmospheric observations and systematic errors cancel each other. Mean ^{222}Rn exhalation rates from soils of different textures vary between 30 and 80 Bq m^{-2} h^{-1} [*Schüssler*, 1996]. Our calculation of the mean ^{222}Rn exhalation rate in the catchment area of Schauinsland is based on the actual texture class distribution and has probably an uncertainty of less than ±10 Bq m^{-2} h^{-1} (or ±16%). An additional error of about 7% is contributed from the decay correction, which we assumed to be constant for all air masses (Section 3.4). The total uncertainty of the radon-derived CO_2 flux estimate associated with the ^{222}Rn flux and atmospheric activity determination is therefore between 20% and 25%.

[25] Taking into account this ^{222}Rn uncertainty and possible uncertainties of the CO_2 offset between MBL and Schauinsland station which varies between ±0.71 ppm (1980s) and ±0.54 ppm (1990s) the uncertainty in the monthly mean fluxes were calculated and added as shaded areas in Figures 4a and 4c. Note that the uncertainty of the fossil fuel CO_2 correction of about 10% is negligible compared to the other errors discussed above.

3.7. Comparison of the Radon-Derived Seasonal Biogenic Flux With Model Estimates

[26] Figure 4c shows the monthly mean biogenic net CO_2 flux for the catchment area of Schauinsland station for the time period of 1990 to 1995 in comparison with model results for the same period from the SIB model [*Sellers et al.*, 1996], as well as from the model used by Peylin et al. [*Peylin et al.*, 2000]. The radon-derived results are in surprisingly good agreement with the model results, also considering the uncertainty of the model estimates as well as uncertainties in the ^{222}Rn tracer method. This is very promising, as our approach now opens the possibility of using regional CO_2 and ^{222}Rn observations to calculate the magnitude and changes in biogenic CO_2 fluxes which have to be taken into account when reliably quantifying the carbon balance and its long-term and interannual changes over Europe.

4. Conclusions

[27] We demonstrated that continuous CO_2 measurements in combination with ^{222}Rn observations at continental sites can be used to derive quantitative information on the strength and variability of continental CO_2 sources and sinks. With a rigorous data selection of atmospheric CO_2 measurements based on wind speed and time of the day, we were able to derive a CO_2 concentration record for the Schauinsland station most probably representative for a catchment area on the order of several hundred thousand square kilometers (i.e., southwest Germany and France). Then using the differences of CO_2 mixing ratios between the selected Schauinsland record and the marine background in the respective latitude belt in combination with ^{222}Rn observations and the ^{222}Rn tracer method allowed us to estimate continental CO_2 fluxes.

[28] The mean seasonal cycle of the radon-derived biogenic flux compares very well with estimates from a process-based biosphere model as well as with an inverse model approach for the study region. Annual mean emission rates turned out to show a huge interannual variability of more than a factor of two most probably caused by changes in biogenic fluxes during the vegetation period. The year-to-year variability of the net annual biogenic flux derived from our observations, changing from about −400 tC km^{-2} yr^{-1} in 1986 to +300 tC km^{-2} yr^{-1} in 1998 is a matter of concern when it comes to verification of regional CO_2 emissions. Our analysis shows that this variability in the regional carbon balance lies well in the range of the actual reduction target of 8% of anthropogenic CO_2 emissions proposed for the European Union (mean fossil fuel CO_2 emissions in the catchment area in the observation period are 166 tC km^{-2} $year^{-1}$). Looking at only short periods of verification of reduction targets, i.e., a few years, can thus be very misleading and only atmospheric observations allow us to monitor their long-term trends and anomalies.

[29] Our net CO_2 flux estimates strongly depend on the accuracy to which the monthly mean ^{222}Rn exhalation rate is known and to the accuracy of the statistical data used to reconstruct the fossil CO_2 emissions. Hence a more detailed study of the ^{222}Rn exhalation in Europe will largely improve the quality of flux balances presented here. It should also be kept in mind that the CO_2 gradients between the Schauinsland site and the marine boundary layer which were used in this study to derive flux estimates are on the order of very few ppm. In order to derive a 10% or better accuracy in flux estimates, this requires very high measurement accuracy and intercomparability to better than 0.1 ppm within the international European and global observational network.

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R. Graul, Umweltbundesamt, Messstelle Schauinsland, Postfach 1229, D-79602 Kirchzarten, Germany.

I. Levin, Institut für Umweltphysik, Universität Heidelberg, INF 229, D-69120 Heidelberg, Germany.

H. Sartorius, Bundesamt für Strahlenschutz, IAR, Rosastrasse 9, D-79098 Freiburg, Germany.

M. Schmidt, LSCE, CE Saclay, Bât. 709, Orme des Merisiers, F-91191, Gif-sur-Yvette Cedex, France.