Can the envisaged reductions of fossil fuel CO\(_2\) emissions be detected by atmospheric observations?

Ingeborg Levin · Christian Rödenbeck

Abstract The lower troposphere is an excellent receptacle, which integrates anthropogenic greenhouse gases emissions over large areas. Therefore, atmospheric concentration observations over populated regions would provide the ultimate proof if sustained emissions changes have occurred. The most important anthropogenic greenhouse gas, carbon dioxide (CO\(_2\)), also shows large natural concentration variations, which need to be disentangled from anthropogenic signals to assess changes in associated emissions. This is in principle possible for the fossil fuel CO\(_2\) component (FFCO\(_2\)) by high-precision radiocarbon (\(^{14}\)C) analyses because FFCO\(_2\) is free of radiocarbon. Long-term observations of \(^{14}\)CO\(_2\) conducted at two sites in southwestern Germany do not yet reveal any significant trends in the regional fossil fuel CO\(_2\) component. We rather observe strong inter-annual variations, which are largely imprinted by changes of atmospheric transport as supported by dedicated transport model simulations of fossil fuel CO\(_2\). In this paper, we show that, depending on the remoteness of the site, changes of about 7–26% in fossil fuel emissions in respective catchment areas could be detected with confidence by high-precision atmospheric \(^{14}\)CO\(_2\) measurements when comparing 5-year averages if these inter-annual variations were taken into account. This perspective constitutes the urgently needed tool for validation of fossil fuel CO\(_2\) emissions changes in the framework of the Kyoto protocol and successive climate initiatives.

Keywords Carbon dioxide · Radiocarbon analyses · Kyoto protocol

Introduction

The European Union (EU) has committed itself under the Kyoto protocol (2007) to reduce its greenhouse gases emissions to 92% of the emissions in 1990. This reduction must be achieved by year 2012, calculated as average over the 5 years commitment period (2008 to 2012). Very recent negotiations even yielded reduction targets in the EU of 20% (International Herald Tribune 2007). The United Nations Framework Convention on Climate Change (UNFCCC) has obliged all participating countries to regularly report on their anthropogenic emissions (and removals of sinks) of all greenhouse gases, in particular carbon dioxide, using comparable methodologies agreed upon by the Conference of Parties. Given these requirements, there is an urgent need for reliable monitoring and regular assessment of greenhouse gases emissions, as ratification of the Kyoto protocol is legally binding.

However, a yet unresolved problem is to actually find an appropriate tool to regularly assess reported emissions. In particular, with respect to the implications on climate change, it is not sufficient to estimate emissions at the bottom-up level (i.e., from statistics of fuel consumption and emissions factors), as these could be significantly biased if relevant sources are missing. The ultimate proof of emissions changes (reductions) is their manifestation in the atmosphere.
atmosphere, via actually observed changes of respective mixing ratios. This requires precise long-term atmospheric observations (Nisbet 2005). In contrast to stratospheric ozone depleting trace substances (banned by the Montreal Protocol), where the overwhelming effect is directly visible and quantifiable by world-wide decreasing or at least stabilizing atmospheric mixing ratios of the long-lived chlorofluorocarbons (Montzka et al. 1999; Prinn et al. 2000; unpublished data from the Advanced Global Atmospheric Gases Experiment (AGAGE)), the case of carbon dioxide (as well as methane and nitrous oxide) is not so simple: Here, the anthropogenic “disturbance” is superimposed on relatively large and strongly variable natural fluxes of CO₂ between the atmosphere, the terrestrial biosphere, and the oceans (Bousquet et al. 2000; Rödenbeck et al. 2003; Geels et al. 2006). In the last decades, about half of the global CO₂ emissions from fossil fuel burning have been taken up by the continental biosphere and the world oceans (Denman et al. 2007), with the remaining half causing the observed increase of approximately half a percent per year in the global atmosphere (GLOBALVIEW 2006). It can, however, not be warranted that this partitioning will persist into the future.

In the Kyoto context, the changes in regional fossil fuel emissions are relevant. These changes lead to proportional changes in the regional excess fossil fuel CO₂ component (ΔFFCO₂) in the atmosphere. ΔFFCO₂ denotes the difference between the fossil-fuel-related CO₂ load in the polluted atmosphere and that in background air. This regional excess fossil fuel CO₂ component can be specifically derived from high-precision atmospheric ¹⁴CO₂ observations (Levin et al. 1989, 2003). All living biomass and dissolved carbon in the oceans contain a certain amount of ¹⁴C, which is received from CO₂ exchange with the atmosphere. In contrast, fossil fuels have been disconnected from this exchange since hundreds of millions of years so that any ¹⁴C originally present in fossil fuels has decayed (the radioactive half life of ¹⁴C is 5,370 years). Thus, over highly populated areas with large CO₂ emissions from fossil fuel burning, such as Central Europe, we are, in principle, able to estimate the regional fossil fuel CO₂ burden from measurements of ¹⁴C in atmospheric CO₂: Comparing the ¹⁴CO₂ level at a polluted site with respective measurements in background air of the same latitude, any depletion of the ¹⁴C/C ratio of CO₂ at the polluted site can be directly translated into a regional excess fossil fuel CO₂ mixing ratio (see below for details). As even over highly populated regions such as Europe, this FFCO₂ excess is generally rather small (only a few ppm); very precise observations of ¹⁴CO₂ are essential to apply this method, making these data still very sparse.

Materials and methods

In Germany, precise long-term ¹⁴CO₂ measurements have been conducted for more than 20 years at two sites: (1) at the Schauinsland station located at 1205 m a.s.l. in the Black Forest (south-western Germany) and (2) in the suburbs of Heidelberg in the highly populated and polluted upper Rhine valley (Fig. 1a,b; Levin et al. 2003, 2007). CO₂ samples have been collected over 2 weeks and high-precision ¹⁴C analysis made by conventional counting technique (Levin et al. 1980; Kromer and Münnich 1992).
To estimate regional fossil fuel CO₂ from measured ¹⁴CO₂ and CO₂ mixing ratios, we use the following mass balance equations:

\[
\text{CO}_2\text{meas} = \text{CO}_2\text{bg} + \text{CO}_2\text{bio} + \text{CO}_2\text{foss}
\]

(1)

\[
\text{CO}_2\text{meas} (\Delta^{14} \text{C}_{\text{meas}} + 1000 \delta) = \text{CO}_2\text{bg} (\Delta^{14} \text{C}_{\text{bg}} + 1000 \delta) \\
+ \text{CO}_2\text{bio} (\Delta^{14} \text{C}_{\text{bio}} + 1000 \delta) + \text{CO}_2\text{foss} (\Delta^{14} \text{C}_{\text{foss}} + 1000 \delta).
\]

(2)

In this equation, CO₂meas is the observed CO₂ mixing ratio at the polluted continental site, CO₂bg the mixing ratio in the free troposphere (here taken from GLOBALVIEW 2006), CO₂bio the regional biogenic component, and CO₂foss the regional fossil fuel component. The ¹⁴C/C ratios of these components in the Δ-notation are respectively Δ¹⁴C_meas, Δ¹⁴C_bg, Δ¹⁴C_bio, and Δ¹⁴C_foss. Δ¹⁴C is the δ-deviation of the ¹⁴C/C ratio from the NBS Oxalic Acid standard activity corrected for decay (Stuiver and Polach 1977). Solving for CO₂foss yields:

\[
\text{CO}_2\text{foss} = \frac{\text{CO}_2\text{meas} (\Delta^{14} \text{C}_{\text{meas}} - \Delta^{14} \text{C}_{\text{bio}}) - \text{CO}_2\text{meas} (\Delta^{14} \text{C}_{\text{meas}} - \Delta^{14} \text{C}_{\text{foss}})}{\Delta^{14} \text{C}_{\text{bio}} + 1000 \delta}.
\]

(3)

Contrary to the method used by Levin et al. (2003), Eq. 3 explicitly takes into account that the biospheric component may be slightly different in Δ¹⁴C from the atmospheric ¹⁴CO₂ background level (Turnbull et al. 2006). As background Δ¹⁴C, we used a fitted curve through our measurements performed at the High Alpine Research station Jungfraujoch at 3,450 m a.s.l. in the Swiss Alps (Levin and Kromer 2004; Levin et al. 2007; solid sinusoidal line in Fig. 1a,b). For an estimate of the temporal development of Δ¹⁴C_bio, we used model calculations made by Naegler (2005) of the biogenic heterotrophic respiration in mid-latitudes of the northern hemisphere. This heterotrophic respiration represents only about half of the biogenic CO₂ component observed in Heidelberg. The other half from autotrophic respiration of plants was approximated by atmospheric background Δ¹⁴C_bg. With these assumptions, Δ¹⁴C_bio changes from 205‰ in mid-1986 to 80‰ in mid-2006.

In November 2000, the Heidelberg sampling site has moved from the roof of the old Institute building (INF 366) to a new building (INF 229) about 500 m further to the East. For an overlapping period of more than 1 year, samples were therefore collected at both sites to allow for adjustment of the two records and establishment of one consistent long-term sequence. This small adjustment of ΔFFCO₂ = +6.4%) for the measurements at INF 366 is described in detail by Levin et al. (2007). The correction for the influence of a nearby nuclear power plant (Philippsburg) was made as in Levin et al. (2003).

From the ¹⁴C measurement error of individual 2-weekly integrated samples of Δ¹⁴C_meas = ±2–3‰, an uncertainty of the ¹⁴C background level (of about ±1‰) and of ¹⁴C_bio (of less than ±10‰), we can estimate an uncertainty of the monthly mean excess fossil fuel CO₂ for Schauinsland station to less than ±1.0 ppm. For Heidelberg, we have to take into account the additional uncertainty of the nuclear power plant correction and a slightly larger measurement error for all samples analyzed before 2001, so monthly mean ΔFFCO₂ can be determined here as better than ±1.5 ppm. Annual mean ΔFFCO₂ values have thus an uncertainty of better than 0.3 ppm at Schauinsland and of better than 0.45 ppm in Heidelberg (Levin et al. 2007; for later use, these and subsequent numbers are collected in Table 1).

**Results**

Figure 1c shows the ¹⁴C-based monthly mean excess fossil fuel CO₂ at Schauinsland and Heidelberg relative to Jungfraujoch for the whole period of observations. The annual means of these regional FFCO₂ excess mixing ratios are presented in Fig. 1d for Schauinsland and Fig. 1e for Heidelberg. The long-term mean of the regional FFCO₂ excess (and its standard error) at Schauinsland amounts to 1.31±0.09 ppm, while on average, 10.96±0.20 ppm FFCO₂ excess is observed at the polluted Heidelberg site. But more important in the context of the Kyoto protocol and its reduction target is that, neither in Heidelberg nor at Schauinsland, we can detect a significant decreasing (or increasing) trend of the fossil fuel CO₂ component yet. For example, a mean ΔFFCO₂ of 1.41±0.15 ppm at Schauinsland (11.09±0.24 ppm in Heidelberg) is calculated for the first decade of observations, while in the second decade, the mean FFCO₂ excess was 1.27±0.13 ppm for Schauinsland.

| Table 1 | Summary of statistical properties of the yearly ΔFFCO₂ time series shown in Fig. 1d,e, together with estimated measurement uncertainty of yearly values |
|---------|--------------------------------------------------|
|         | Schauinsland | Heidelberg |
| Observed mean | 1.31±0.09 ppm | 10.96±0.20 ppm |
| Observed year-to-year standard deviation | 0.40 ppm | 0.90 ppm |
| Modeled year-to-year standard deviation | 0.26 ppm | 0.65 ppm |
| Measurement uncertainty | <0.30 ppm | <0.45 ppm |
(10.92±0.34 ppm in Heidelberg). A missing trend is, in fact, not surprising when looking at the emissions trends in the area of influence, which for both sites is mainly southern Germany and France (Fig. 2). Although in Germany total reported emissions have decreased since 1990 by more than 10% (UNFCCC 2007), the emissions both in southern Germany and in France have slightly increased or stayed constant (Statistisches Landesamt Baden-Württemberg 2007; UNFCCC 2007). In the closer area of influence of the Heidelberg site, reported fossil fuel CO\(_2\) emissions stayed practically constant [in the Rhein-Neckar area, they increased by about 1% (Statistisches Landesamt Baden-Württemberg 2007)].

We do, however, observe considerable year-to-year variations of the regional excess fossil fuel CO\(_2\) component. The standard deviation of these variations at Schauinsland is 0.4 ppm (31% of the long-term mean, see Table 1) and at Heidelberg is 0.9 ppm (8% of the long-term mean). At least three possible reasons can be made responsible for this observed variability: (1) limited precision of the \(^{14}\)C measurements and validity of the assumptions made to estimate the regional FFCO\(_2\) excess from \(^{14}\)CO\(_2\) observations, (2) temporal variability of the FFCO\(_2\) emissions in the area of influence of the site, and (3) variability of atmospheric transport. The analytical uncertainty of the \(^{14}\)C measurements at Schauinsland, the natural variability of the \(^{14}\)CO\(_2\) background at Jungfraujoch, and the correction for biogenic \(^{14}\)C emissions, as discussed above, can explain about half of the year-to-year variability of \(\Delta\)FFCO\(_2\) (Table 1). Reported inter-annual variations of emissions are small, in the order of 4% at most (UNFCCC 2007). Thus, variable atmospheric transport (changes of the areas of influence of the sites and of the dilution of source CO\(_2\)) is likely to significantly contribute to the observed inter-annual \(\Delta\)FFCO\(_2\) variability.

To test this hypothesis, we performed a dedicated long-term model run from 1986 to 2006 with the global atmospheric transport model TM3 (Heimann 1996), simulating \(\Delta\)FFCO\(_2\) at Schauinsland and Heidelberg. The model transported temporally constant fossil fuel CO\(_2\) emissions as reported by the EDGAR data base (Olivier et al. 2005) on a 1°×1° grid for the year 2000 (for the spatial distribution of emissions, see Fig. 2). The horizontal resolution of TM3 was 1.8°×1.8°. Meteorological driving fields were taken from the US National Centers for Environmental Prediction (NCEP) re-analyses for the time span in question (Kalnay et al. 1996). Monthly mean TM3 FFCO\(_2\) results for Jungfraujoch, smoothed with a harmonic fit as that used for the observations, were subtracted from the Schauinsland and Heidelberg monthly mean model estimates. Yearly means of the resulting modeled excess fossil fuel CO\(_2\) values are plotted in comparison with the observations in Fig. 1d,e. In these plots, constant offsets of −2.0 ppm for Schauinsland and +0.9 ppm for Heidelberg have been added to the simulations so as to match the observed long-term means, to highlight the temporal variations.

Similar to the observations, there is considerable inter-annual variability found in the model estimates. As constant FFCO\(_2\) fluxes have been used for the whole period while the meteorological fields vary from year to year, this modeled variability is solely due to atmospheric transport. The standard deviation of the modeled annual mean \(\Delta\)FFCO\(_2\) is 0.26 ppm at Schauinsland and 0.65 ppm in Heidelberg. Importantly, there is considerable agreement between model and observations in the temporal patterns at

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**Fig. 2** Fossil fuel CO\(_2\) emissions and influence areas of the Schauinsland and Heidelberg measurement sites. **Background color:** Fossil fuel emissions from each 1°×1° pixel of the EDGAR inventory for the year 2000 given in kgC m\(^{-2}\) year\(^{-1}\) (Olivier et al. 2005). **Isolines:** Relative strength of the influence of a local surface flux on the atmospheric mixing ratio at Schauinsland (a) or Heidelberg (b). The plots show the change in yearly concentration at the measurement location that would result from a unit change in the yearly surface emission at a considered location, as calculated by the adjoint TM3 transport model on 1.8°×1.8° spatial resolution. The isolines (counted from outside) indicate 20, 40, 60, or 80% of the maximum value obtained next to the respective station. The shifts between the actual station locations (asterisks) and the apparent centers of influence result from the limited model resolution.
both sites. All major positive or negative excursions from the long-term mean line up between the two-time series. In addition, there are similarities in persistent trends of mixing ratios, such as the increase of $\Delta$FFCO$_2$ by almost 1 ppm at Schauinsland from 1998 to 2006.

Due to the coarse spatial resolution of the global transport model TM3 used here, the simulations cannot be expected to accurately represent the CO$_2$ signals at the Schauinsland mountain station or the urban site Heidelberg, both having very heterogeneous environments in terms of atmospheric circulation and CO$_2$ source patterns. Unfortunately, it is generally very difficult to estimate the uncertainty of transport model results, which can be large (e.g., Geels et al. 2006). Therefore, the model results presented in this paper are mainly used in a qualitative sense. (One may assume that temporal variability can be simulated with some confidence if it is determined by changes in regional transport pathways. In contrast, long-term values may be more affected by systematic errors, which provides a partial justification for adding long-term offsets to our model time series in Fig. 1d,e).

Despite these model limitations, however, the similarity of the observed annual $\Delta$FFCO$_2$ time series to that modeled from constant fluxes does point to variable atmospheric transport being responsible for a significant part of the total $\Delta$FFCO$_2$ variability. (This seems to be the case not only for the changes from 1 year to the next, but also for the before-mentioned recent trend at Schauinsland: As there is no such increase of fossil fuel emissions in the catchment area of the Schauinsland site, this trend must be due to a long-term change of the transport pattern and possible associated changes of the area of influence of this site). To the extent that the values summarized in Table 1 are realistic, transport-related variability is of similar magnitude as the estimated measurement uncertainty (at the polluted site Heidelberg even slightly larger). Further, both contributions add up, in a quadratic sense, to the observed standard deviation of the scatter relative to the mean (as read from Table 1, first two lines) and the available number of yearly values in the two periods to be compared. For a comparison between a 20-year “present” period and a 5-year commitment period (e.g., 2008–2012), the $t$-test yields that changes must be larger than about 26% in the catchment area of the rural slightly polluted mountain site Schauinsland to be detected by our method at the 95% confidence level (assuming equal standard deviations in both periods, one-sided test). At the more polluted urban site Heidelberg, we will be able to already detect changes of about 7%. The corresponding reductions with only 5 years “reference” and 5 years “commitment” periods would have to be larger than 36% at Schauinsland and 10% in Heidelberg.

The situation can be improved if we assume that the transport-related variability of the observed FFCO$_2$ excess can, at least partially, be accounted for by a suitable model framework. This would involve inverse techniques to estimate changes in emission strengths, combining atmospheric data and bottom-up information, and preferably a transport model of higher resolution than the one used in this study (if computationally feasible). To anticipate the effect of such a framework, we assume here that one can create a “transport-corrected” series of annual $\Delta$FFCO$_2$ values whose remaining variability largely represents model and measurement uncertainties. From the correlation of the observed year-to-year variations with those simulated even by our coarse model, we qualitatively conclude that this is sufficiently realistic. Based on the results above, we assume that the standard deviation of these “transport-corrected” time series would drop by a factor around $\sqrt{1/2}$ with respect to the observed time series. The thresholds of detectable changes for a 5-year reference and a 5-year commitment period would then decrease by the same factor, i.e., to about 26% at a site like Schauinsland and about 7% at a polluted site like Heidelberg.

Although these examples can only give an indication, they do show that, depending on the character of the site, fossil fuel CO$_2$ emission reduction targets of 8 or 20% could indeed be confronted with an independent top-down monitoring and verification tool, provided that precise and long-term atmospheric $^{14}$CO$_2$ observations would be available for the area of concern. Measurements would be necessary at sufficiently many sites as determined by the spatial heterogeneity of emission changes and the catchment areas of the measurement locations. At sites with a very small regional fossil fuel CO$_2$ component, the period for verification may, however, need to be quite long. Work is also needed to assess (and improve) the ability of

Discussion

What does this imply for the question of this paper: Can the envisaged reductions of fossil fuel CO$_2$ emissions be detected by atmospheric observations? To answer this on the basis of the existing measurements, we assume that the $\Delta$FFCO$_2$ time series consist of statistically independent and Gaussian-distributed values around a mean value that is representative for the fossil fuel emissions within the given period. Then, we can use a statistical $t$-test to determine the minimum difference between the mean values of two such time series that would be detectable with statistical significance in the presence of the scatter, given the standard deviation of the scatter relative to the mean (as read from Table 1, first two lines) and the available number of yearly values in the two periods to be compared. For a comparison between a 20-year “present” period and a 5-year commitment period (e.g., 2008–2012), the $t$-test yields that changes must be larger than about 26% in the catchment area of the rural slightly polluted mountain site Schauinsland to be detected by our method at the 95% confidence level (assuming equal standard deviations in both periods, one-sided test). At the more polluted urban site Heidelberg, we will be able to already detect changes of about 7%. The corresponding reductions with only 5 years “reference” and 5 years “commitment” periods would have to be larger than 36% at Schauinsland and 10% in Heidelberg.

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available transport models for the present purpose. Our findings have also implications for the interpretation of non-fossil fuel atmospheric CO₂ (as well as other greenhouse gases) records over continents in general: Due to large inter-annual variations in natural fluxes, the statistically significant determination of trends is expected to be more difficult than for the fossil fuel emissions.

In summary, long-term high-precision atmospheric ¹⁴C observations at two monitoring stations in south-western Germany have been used to estimate changes in regional fossil fuel emissions. To date, no significant changes have been revealed yet, in accordance with the emission inventories for the estimated regions of influence. However, it could be shown that politically envisaged changes of 8 or 20% could be detected, provided that long enough time series exists in a dedicated observational network. Such a network must be selected by appropriate sensitivity studies and supported by high-resolution atmospheric transport models. Respective observations, monitoring the relevant parts of Europe and elsewhere on the globe, do, however, have to start soon; otherwise, this powerful tool will come too late.

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