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ORIGINAL PAPER

Can the envisaged reductions of fossil fuel CO_2 emissions be detected by atmospheric observations?

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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

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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, 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 CO_2 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 CO_2 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 CO_2 component* ($\Delta FFCO_2$) in the atmosphere. $\Delta FFCO_2$ denotes the difference between the fossil-fuel-related CO_2 load in the polluted atmosphere and that in background air. This regional excess fossil fuel CO_2 component can be specifically derived from high-precision atmospheric $^{14}CO_2$ observations (Levin et al. 1989, 2003). All living biomass and dissolved carbon in the oceans contain a certain amount of ^{14}C, which is received from CO_2 exchange with the atmosphere. In contrast, fossil fuels have been disconnected from this exchange since hundreds of millions of years so that any ^{14}C originally present in fossil fuels has decayed (the radioactive half life of ^{14}C is 5,370 years). Thus, over highly populated areas with large CO_2 emissions from fossil fuel burning, such as Central Europe, we are, in principle, able to estimate the regional fossil fuel CO_2 burden from measurements of ^{14}C in atmospheric CO_2: Comparing the $^{14}CO_2$ level at a polluted site with respective measurements in background air of the same latitude, any depletion of the $^{14}C/C$ ratio of CO_2 at the polluted site can be directly translated into a regional excess fossil fuel CO_2 mixing ratio (see below for details). As even over highly populated regions such as Europe, this $FFCO_2$ excess is generally rather small (only a few ppm); very precise observations of $^{14}CO_2$ are essential to apply this method, making these data still very sparse.

Materials and methods

In Germany, precise long-term $^{14}CO_2$ 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). CO_2 samples have been collected over 2 weeks and high-precision ^{14}C analysis made by conventional counting technique (Levin et al. 1980; Kromer and Münnich 1992).

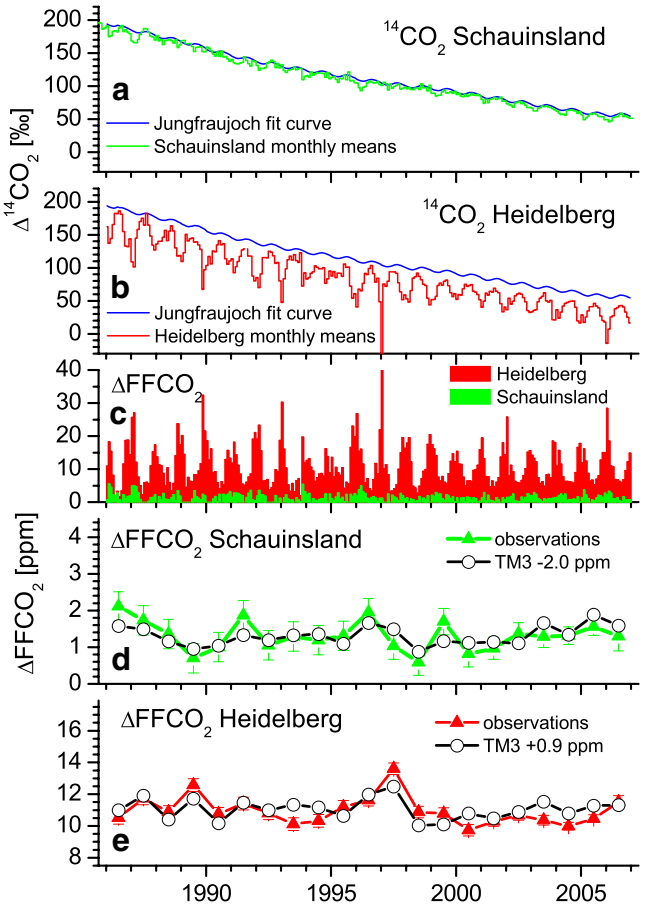

Fig. 1 Long-term observations of $^{14}CO_2$ in south-western Germany and respective ^{14}C-based fossil fuel CO_2 excess compared to Jungfraujoch: **a** Monthly mean observed $^{14}C/C$ ratio in atmospheric CO_2 at Schauinsland and **b** Heidelberg in comparison to the continental reference level derived from respective observations at Jungfraujoch. The long-term decrease at all three stations is due to still ongoing equilibration of the atmospheric bomb $^{14}CO_2$ spike with the ocean and the biosphere and to a steadily increasing fossil fuel CO_2 burden in the global atmosphere. **c** ^{14}C-based monthly mean $\Delta FFCO_2$ at Schauinsland and Heidelberg calculated with Eq. 3. **d** Comparison of annual mean $\Delta FFCO_2$ observed at Schauinsland and **e** Heidelberg with TM3 model estimates. For comparison of the inter-annual variability of model estimates and observations, the model results were shifted (see legend) so that the respective long-term mean values agreed with those of the observations. Errors in the observations are standard mean errors

To estimate regional fossil fuel CO_2 from measured $^{14}CO_2$ and CO_2 mixing ratios, we use the following mass balance equations:

$$CO_{2meas} = CO_{2bg} + CO_{2bio} + CO_{2foss} \quad (1)$$

$$CO_{2meas}\,(\Delta^{14}C_{meas}+1000‰) = CO_{2bg}\,(\Delta^{14}C_{bg}+1000‰) + CO_{2bio}\,(\Delta^{14}C_{bio}+1000‰) + CO_{2foss}\,(\Delta^{14}C_{foss}+1000‰). \quad (2)$$

In this equation, CO_{2meas} is the observed CO_2 mixing ratio at the polluted continental site, CO_{2bg} the mixing ratio in the free troposphere (here taken from GLOBALVIEW 2006), CO_{2bio} the regional biogenic component, and CO_{2foss} the regional fossil fuel component. The $^{14}C/C$ ratios of these components in the Δ-notation are respectively $\Delta^{14}C_{meas}$, $\Delta^{14}C_{bg}$, $\Delta^{14}C_{bio}$, and $\Delta^{14}C_{foss}$. $\Delta^{14}C$ is the ‰-deviation of the $^{14}C/C$ ratio from the NBS Oxalic Acid standard activity corrected for decay (Stuiver and Polach 1977). Solving for CO_{2foss} yields:

$$CO_{2foss} = \frac{CO_{2bg}\left(\Delta^{14}C_{bg} - \Delta^{14}C_{bio}\right) - CO_{2meas}\left(\Delta^{14}C_{meas} - \Delta^{14}C_{bio}\right)}{\Delta^{14}C_{bio} + 1000‰}. \quad (3)$$

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 $\Delta^{14}C$ from the atmospheric $^{14}CO_2$ background level (Turnbull et al. 2006). As background $\Delta^{14}C$, 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 $\Delta^{14}C_{bio}$, 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 CO_2 component observed in Heidelberg. The other half from autotrophic respiration of plants was approximated by atmospheric background $\Delta^{14}C_{bg}$. With these assumptions, $\Delta^{14}C_{bio}$ 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 $\Delta FFCO_2$=+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 ^{14}C measurement error of individual 2-weekly integrated samples of $\Delta^{14}C_{meas}$=±2–3‰, an uncertainty of the ^{14}C background level (of about ±1‰) and of $^{14}C_{bio}$ (of less than ±10‰), we can estimate an uncertainty of the monthly mean excess fossil fuel CO_2 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 $\Delta FFCO_2$ can be determined here as better than ±1.5 ppm. Annual mean $\Delta FFCO_2$ 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 ^{14}C-based monthly mean excess fossil fuel CO_2 at Schauinsland and Heidelberg relative to Jungfraujoch for the whole period of observations. The annual means of these regional $FFCO_2$ excess mixing ratios are presented in Fig. 1d for Schauinsland and Fig. 1e for Heidelberg. The long-term mean of the regional $FFCO_2$ excess (and its standard error) at Schauinsland amounts to 1.31±0.09 ppm, while on average, 10.96±0.20 ppm $FFCO_2$ 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 CO_2 component yet. For example, a mean $\Delta FFCO_2$ 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 $FFCO_2$ excess was 1.27±0.13 ppm for Schauinsland

Table 1 Summary of statistical properties of the yearly $\Delta FFCO_2$ 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

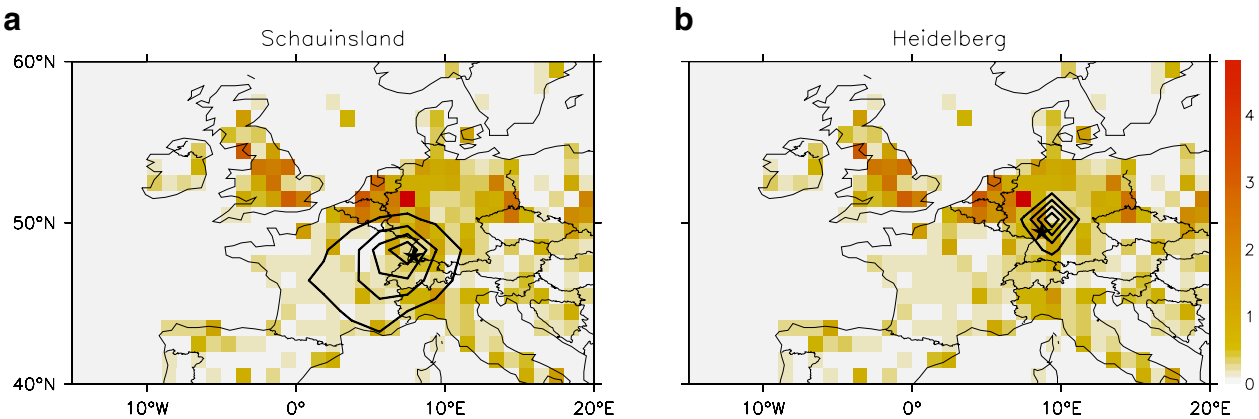

Fig. 2 Fossil fuel CO_2 emissions and influence areas of the Schauinsland and Heidelberg measurement sites. *Background color*: Fossil fuel emissions from each 1°×1° pixel of the EDGAR inventory for the year 2000 given in kgC m^{-2} $year^{-1}$ (Olivier et al. 2005). *Isolines*: Relative strength of 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 surprisingly well. From this finding, we tentatively draw the quantitative conclusion that transport-related variability contributes roughly one half to the year-to-year variability of the observed $\Delta FFCO_2$ time series.

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.

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

available transport models for the present purpose. Our findings have also implications for the interpretation of non-fossil fuel atmospheric CO_2 (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 ^{14}C 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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