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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simulating the integrated $\Delta^{14}CO_2$ signature from anthropogenic emissions over Western Europe" *by* D. Bozhinova et al.

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We thank Dr Turnbull for the constructive and insightful comments that helped us to improve our manuscript. Below we will respond to them point by point.

General comments: Clearly, nuclear industry emissions are an important influence on $\Delta^{14}CO_2$ *in Europe, particularly for Britain and France. However, when fossil fuel* CO_2 *is calculated, as in section 2.1, the choice of background plays a critical role. When a regional background value (e.g. Jungfraujoch) is used, then the nuclear industry emissions must be accounted for explicitly as an additional emission term. But judicious choice of a local background between the nuclear emission source and the region of interest may allow the nuclear industry emissions to be accounted for in the background*

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term instead. If these background measurements are made at the right times/locations, then the nuclear industry emissions can potentially be correctly accounted for, even if these emissions are not well-known (since the nuclear industry impact should be consistent between the background and observed samples). One would imagine that the sampling strategy would be important in this case -flask samples that are collected under a single wind regime would presumably be easier than integrated (plant or absorption) samples. This could be assessed with the existing model runs, and it would be a very useful addition to the paper.

Indeed, the choice of background site is crucial for the estimation of the $\Delta^{14}CO_2$. In our case, we evaluated the nuclear and fossil fuel signal reaching Jungfraujoch in our simulations and we found that the signal was negligible. However, if larger signals of anthropogenic emissions or biospheric enrichment are already included at the chosen site, then care must be taken that these are not double-counted when evaluating the signatures at other locations. We agree that we have not elaborated on this issue, even though its large importance, and we have revised our methods, results and discussion sections to include this. To reply to the question, we tried to find a background location that is influenced by the nuclear signal, but not by large fossil signals at a particular location. We tried a location near Cambridge that we show in our new Figure 7. However, in that particular region both nuclear influence and fossil fuel emissions are quite high, and the signals can hardly be separated. We agree with the reviewer that this issue deserves more attention in future studies.

Section 3.4. Direct estimation of fossil fuel CO_2 *emissions. Clearly this will never be a "perfect" way to estimate emissions, since most of the variability is presumably due to real variability in atmospheric transport regimes (and perhaps the spatial extent of emissions from each city?). Yet 30% uncertainty is as good as we get from any atmospheric method at the moment, so this has potential as a "quick and dirty" way to look for large inconsistencies in reported emissions. However, it is important to clearly detail the limitations as well as the advantages of this idea. In particular, less*

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knowledgeable readers may interpret this to think that observed mole fractions can be directly equated to emission rates, which is simply not the case. I found the writing in this section particularly hard to follow. If this section is included in the final version of the paper, it needs to be expanded and rewritten, to clearly state the premise and it's limitations, and make the readability better.

Thank you for these suggestions. Indeed, we agree that this section had poor readability, which led to misunderstanding our original intentions. In fact our idea is precisely to show that a simpler relationship cannot be used. We have now revised the section and we hope that this will improve its message. In more detail, in Section 3.4 we now present the results of comparing a simple box model to the results from the more comprehensive transport-modeling framework.

Specific comments. Pg 30613, lines 14-25. The phrasing of this section seems to imply that correlate tracers are better than ^{14}C *as fossil fuel* CO_2 *detectors. In fact, the varying emission ratios depending on source present a real challenge, and is the main reason why these tracers are not as useful as* ^{14}C*. It is also worth noting that only some of these tracers are co-emitted with fossil fuel* CO_2*, with others having only approximately co-located, but independent, sources.*

Indeed, we have wrongfully asserted that the varying emission ratios are an advantage when dealing with these trace gases. This is now corrected in the text and the additional description of the complications related to indirect proxies is also included.

Pg 30614 lines 19-23. Integrated flask sampling is also done (e.g. Turnbull, J. C., Guenther, D., Karion, A., Sweeney, C., Anderson, E., Andrews, A. E., Kofler, J., Miles, N. L., Newberger, T., Richardson, S. J., and Tans, P. P.: An integrated flask sample collection system for greenhouse gas measurements, Atmospheric Measurement Techniques, 5, 2321-2327, 10.5194/amt-5-2321-2012, 2012). The time period over which the absorption samples are collected should also be discussed here.

Thank you for this reference, a note regarding this technique is now included in the

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manuscript.

Pg 30615, lines 18-28. This text incorrectly implies aircraft sampling is not influenced by stratospheric exchange or disequilibrium fluxes.

Corrected.

Pg 30617, lines 14-25. The biospheric term will bias the results, and if it is excluded, it needs to be carefully and quantitatively justified. This is particularly true when a regional high-altitude background such as JFJ is used. As in my general comment (above) about choice of background, it may be that judicious choice of background can decrease the importance of the biospheric term, and this could be tested by the model.

Indeed, the omission of the biospheric disequilibrium term, especially over the summertime will result in a systematic bias as the enrichment will be constantly missing. We have now included this in the cited paragraph with quantitative estimation from literature.

P 30620, lines 25-27. Please add a figure to show the domain.

We have now included a figure showing our model domains.

Pg 30622, lines 2-3. Absorption samplers do not necessarily sample continuously, they can be controlled to switch on and off (e.g. only under certain wind conditions or during certain time periods – e.g. daytime only), and some labs do this already.

A note of this is added in the paragraph.

Pg 30614, lines 8-12. The model-obs agreement at Schauinsland is not terribly good. As the authors point out, it is hard to compare carefully with only 6 data points. It would be worth comparing this model output with other modeling studies and with any other ^{14}C datasets that are available for Europe. For example, the Turnbull et al. (2009) LMDZ modeling study, runs through to 2007, and has full seasonal cycles for several years. This could be compared with the WRF-CHEM seasonal cycle and spatial variability

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(noting the coarser resolution of the LMDZ model and the simplified nuclear industry source used there). Could the model results also be compared with the Palstra et al (2008) wine ethanol measurements (even though the years are different)?

We prefer to not compare our results from 2008 to another year, because we have noted that the differences in the transport, boundary conditions, anthropogenic emissions and background $\Delta^{14}CO_2$ make the comparison difficult to judge quantitatively. This is true even when correcting for the downward ^{14}C trend. For the current manuscript, we have now included comparison to a few other sites with $\Delta^{14}CO_2$ observations in our new Fig 3. With regard to the comparison of the seasonal cycle, unfortunately our modeled period covers only half the year. As such, it is difficult to evaluate the annual trend contribution and compare with de-trended seasonality.

Pg 30624-30625, lines 17 - 7. This disagreement at Lutjewad is concerning, as a major motivation for moving to regional models such as WRF-CHEM is to improve the model performance at small scales and at individual sites. It would be surprising if there were a measurement problem with these samples, and if there is, it needs to be carefully explained. The model output is presumably available at higher than the monthly resolution shown in the plot, so it could be investigated in more detail to see which components of the ^{14}C flux might be causing the mismatch, and whether this is related to errors in model transport at this location. Conditional sampling based on wind direction is also carried out at Lutjewad, so these samples could be compared with the model to shed more light on this problem.

We investigated the model-to-observations mismatch for the station of Lutjewad in our simulation of summertime 2008 $\Delta^{14}CO_2$ signatures. Our co-authors from Groningen provided us with the longer time series of their 24-hour monthly $\Delta^{14}CO_2$ observations (parts of which are unpublished yet) and we were able to evaluate the seasonal cycle beyond the year of our study. More specifically, we used the NOAA Earth System Research Laboratory's ccgcrv routine (*Thoning and Tans*, 1989) to fit the data. We show the de-trended seasonal cycle below. It is immediately visible that 2008 is a

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special year for this location as it showed considerably lower signatures during the entire year (Reply-Fig. 1), a decrease in the long-term trend, and atypical seasonality with a missing summer peak (better seen on Reply-Fig. 2). As we are unsure what exactly is the reason for the anomalous behavior it is difficult to evaluate why our model is not capturing this in 2008. We do notice that a similar year with a lacking summer maximum occurs in 2012, suggesting that the 2008 observations are not necessarily suspicious from the measurement technical point of view. Our Groningen co-authors have confirmed that in principle they see no measurement related problems with the data presented, but they also do not yet have a physical explanation for the observed anomaly in the trend and seasonal cycle.

Since we cannot find any obvious flaws in the data, we decided to maintain the Lutjewad time series in the comparison despite the large mismatch. We also included the south sector observational data in new Figure 3. We state in the text that the year 2008 in Lutjewad was different from other years in the long-term record, in ways that are obviously not captured by the model as it produces the more typical seasonal pattern also seen at Jungfraujoch and Schauinsland. Further analysis of this anomalous signal, and the possible model improvements it might yield, are part of ongoing work. In new Figure 4 in the manuscript our graph C) will now focus on showing the modeled high-frequency variations of the signature, without comparison to the monthly integrals.

Pg 30625 – section 3.2. Given the concerns in the previous section about possible errors in the modeled nuclear influence at Lutjewad, how reliable is the interpretation given for the overall nuclear influence? See also my general comment about choice of background to reduce the nuclear influence. In the sentence that is referred we mention that the nuclear signals can be sometimes of the magnitude of the fossil fuel signal at that site. Even then our model results do not suggest considerable nuclear influence at Lutjewad overall. But we will try to answer the second part of the reviewer's question in more general manner.

While the total uncertainty of the nuclear estimates is largely unknown, mostly due to

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the missing information about the temporal variability of the emissions, it is possible to evaluate some uncertainties connected with the method used. We have now included a new Figure 6, which shows the uncertainty in the modeled nuclear $^{14}CO_2$, which is associated with the uncertainty in the emission factors of the different reactor types.

Pg 30628 Section 3.4. See my general comment on this section.

Replied with the general comments.

Pg 30630, lines 5-7. Please revise to clarify what is meant by this sentence.

Done.

Pg 30630, lines 22-24. I'm not sure that it is true that reactor ^{14}C emissions can be expected to be constant. Emissions may be dominated by short maintenance periods or periodic emission episodes – this statement needs to be justified.

We have elaborated and revised this paragraph.

Pg 30631 lines 0-12. When there's a big nuclear influence in a single flask sample, yes, it will be obvious. And as the authors state, such signals will be less evident in integrated (plant or absorption) samples. It is this latter than seems more concerning, since a small signal won't be readily apparent, but will still cause a systematic underestimate of fossil fuel CO_2. A test of the model results of how much this matters would be helpful. I.e. for plant samples, how much bias does the nuclear industry exert?

We have now included a new panel in Figure 9 that we hope answers this question. Generally, the bias from the nuclear emissions for the plant samples that we have modeled is within 0-4 ‰ for the continental part of our domain, exceeding this numbers only in the grid cells containing the emission sources. This is partly because in the continental part of our domain most reactors are of the types of boiling water or pressurized water reactors, which emit lower amounts of ^{14}C. For the territory of the United Kingdom, where there are several gas-cooled reactors, which emit on average more than 10 times higher amounts of ^{14}C, the bias is higher and found further away from the

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sources.

Pg 30631 lines 11-12. Indeed, better characterization of the nuclear emissions is needed, but better strategies to minimize the influence of them when using ^{14}C measurements is also needed – see my general comment.

Indeed, this is now added in our discussion.

Pg 30632 lines 4-9. I am not convinced that plant sampling is likely to be better that integrated absorption samples. It is true that plants only sample during the day, but altering integrated absorption samplers to sample only during the day is an easy fix and some researchers already do this. The plant assimilation weighting function will always be difficult to pin down exactly, whereas absorption sampling times can be controlled easily and recorded.

It is true that altering current sampling techniques is easier and provides less uncertainty. However, the advantage of using plant samples to complement regular observations is that it can provide some spatial resolution in a very scarce observational network.

References

Thoning, K., and P. Tans (1989), Atmospheric carbon dioxide at mauna loa observatory. 2. analysis of the NOAA GMCC data, 1974-1985, *J. Geophys. Res.*, *94*(D6), 8549–8565.

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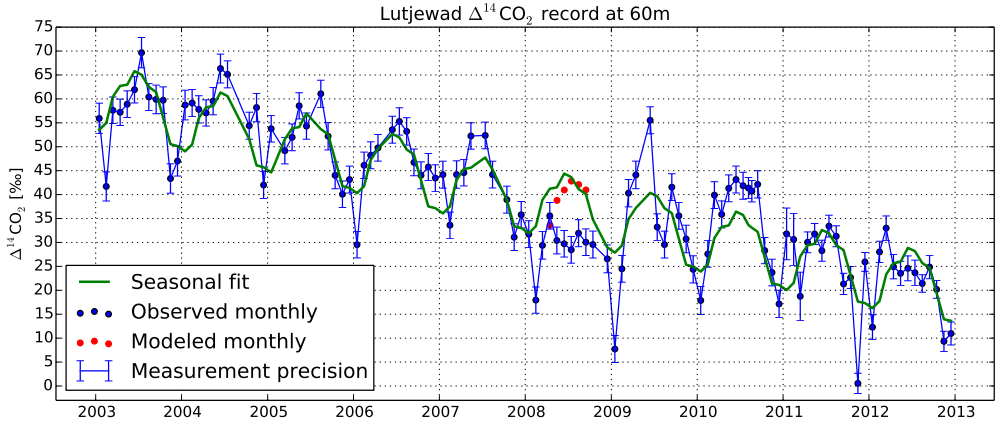

Fig. 1. Lutjewad Δ14CO2 observations and functional fit, in addition to modeled values for 2008 (source of observational data: Sanne Palstra, Centre for Isotope Research, Groningen, personal communication)

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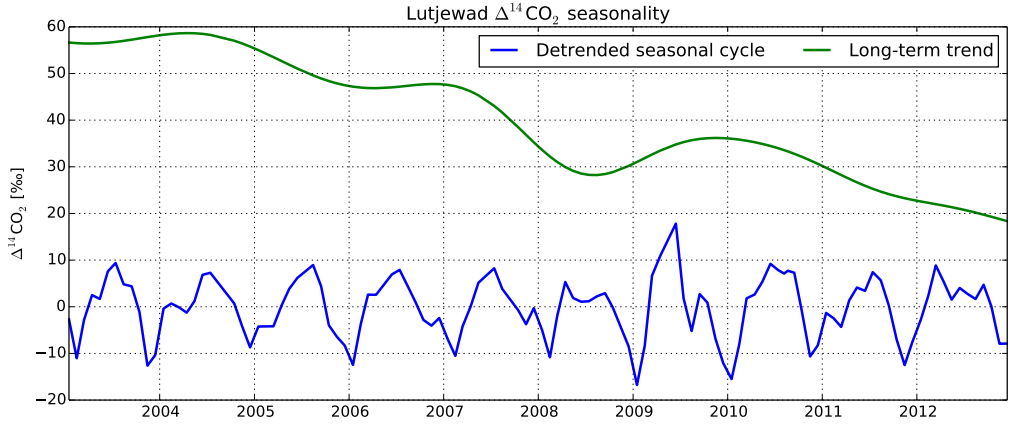

Fig. 2. De-trended seasonal cycle and long-term trend of the Lutjewad Δ14CO2 observations

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