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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 Prof. Dr. Levin for the detailed review of our work and the constructive comments that helped us revise and improve our manuscript. Below, we address all issues raised by the reviewer.

This study includes all known relevant components for its purpose. However, it largely lacks adequate comparison with observations for its evaluation. Much more observational data than only from Lutjewad (NL) are indeed available from the published literature since the 1980s, and also for the year 2008 (e.g. for the station Heidelberg (south west Germany), which is located in a polluted area and is influenced by nuclear ^{14}C emissions, see Levin et al., Phil. Trans. R. Soc. A (2011) 369, 1906–1924). The

latter impact of the nuclear emissions has extensively been monitored from 1983-1985 (Levin et al., Health Physics 54, 149-156, 188). Also for the highly "contaminated" surroundings of the Sellafield reprocessing plant in the UK a large set of ^{14}C observations from plant material exists for the years 1983-1985 (McChartney et al., 1988); these two data sets, although not from the year 2008, may be very illusive, at least for a semi-quantitative comparison with the model simulations.

Following the advice of the reviewer we looked for additional observational data for $\Delta^{14}CO_2$ to compare to our model simulations. Monthly and weekly integrated samples from Heidelberg, Germany, Prague-Bulovka, and Kosetice, Czech Republic are now included in the model-to-observation comparison. Some data from other years is available too, but we prefer not to 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.

Directly comparing our modeled plant signatures to the ones observed near Sellafield as suggested, is even more complicated as the study was conducted more than 30 years ago. Both the atmospheric background $\Delta^{14}CO_2$ was much different at that time, and the signals from the disequilibrium fluxes from ocean and biosphere were larger. Additionally, Sellafield nuclear emissions were considerably higher – in the cited paper they estimate 20 TBq annual emissions, while since the year 2000 this site reports less than 1 TBq per year ^{14}C emissions.

Unfortunately, the comparison with ^{14}C data and CO-based fossil fuel CO_2 estimates from the Lutjewad (NL) station, which is discussed here, is not really convincing. This is also because there are some obvious mis-understandings, which, I suspect, could have been avoided with engagement from co-authors, see comments on Pages 30624-30625 below.

While we discuss the CO-based fossil fuel CO_2 data in a following comment, we fur-

ther 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 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 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 the new Figure 4 C) in the manuscript we will now focus on the modeled high-frequency variations of the signature, without comparison to the monthly integrals.

Besides the above mentioned, I have a number of other points which need revision before publication in ACP: Abstract last sentence: As mentioned above, although I think that the modeling framework presented here is a comprehensive one, I am still

confused by the rather large deviations between modeled and observed components (for details see below).

We have addressed the concerns regarding the model to observation comparisons in other comments and included extra observational data in our study. We hope that with these new additions we have shown that our modeling framework is well suited for this purpose, as the sentence in the abstract states.

Page 30613, lines 18-20: I do not agree that it is an advantage that e.g. the CO/FFCO_2 ratio for traffic emissions is so much different from that of other FFCO_2 sources. This is rather the largest problem we are confronted with when simply using only one mean emission factor for all fossil fuel sources and not "re-calibrating" the CO/FFCO_2 ratio with ^{14}C data on the seasonal and diurnal time scale to account for the temporally changing source mix.

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.

Page 30614, line 1: The half life time of ^{14}C is now revised to 5700$\pm$30 yrs (see: Roberts M.L. und Southon J.R. 2007. "A preliminary determination of the absolute ^{14}C/12C ratio of OX-I". Radiocarbon 49 (2): 441–445).

Thank you for this reference, it has replaced the previously used one.

Page 30614, line 3: Only slightly more than 50% of ^{14}C is produced in the stratosphere, the remaining in the upper troposphere (see Lal, D., and B . Peters: Cosmic ray produced radioactivity on the Earth, in Handbuch der Physik, vol. No. XLVI/2, pp. 551-612, Springer Verlag, New York, 1967, or Masarik and Beer, JGR 104, D10, 12,099-12,111, 1999).

The sentence now includes the "upper troposphere" as well, and we kindly thank you for the references.

Page 30614, lines 17-18: Please note that (at least to my knowledge) there is no ^{14}C

counting technique in use that is measuring graphite targets.

Indeed, we have corrected the wording in the sentence to properly describe both procedures.

Page 30617, line 3: Here a CO_2 term from the stratosphere is missing.

Corrected.

Page 30617, line 23ff: It is not totally clear if all components of the ecosystem respiration $^{14}CO_2$ are really very similar to background air. In Levin et al. (SCIENCE OF THE TOTAL ENVIRONMENT 391 (2008) 211–216) and also in other publications (e.g. van der Laan et al., 2010) a different (i.e. higher) value is assumed.

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: "We should note, though, that the omission of the biospheric disequilibrium in the region and period of our study will likely results in a small bias in our results, which will be less enriched during the period of peak biospheric activity. Turnbull et al. (2006) estimates for the Northern Hemisphere this term to be on average equivalent to the overestimation of fossil fuel CO_2 by 0.2-0.5 ppm or up to 1.3 ‰ enrichment in $\Delta^{14}CO_2$, while Levin et al. (2008) evaluates this influence on the observational sites in Germany to be within 0.2 ppm or about 0.5 ‰ enrichment."

Page 30618, lines 1-3: It should be mentioned here, that e.g. for the Heidelberg fossil fuel CO_2 estimates – i. e. already in Levin et al. (2003) - we made a correction based on measured (!) monthly $^{14}CO_2$ emission data from the NPP Philippsburg and a Gaussian Plume model.

Indeed, the cited study is one of the few that have tried to correct for this influence and is now included in the paragraph.

Page 30618, lines 7-10: The extensive measurement data from the surroundings of

the reprocessing plant in Sellafield (i.e. the data from the paper by McCartney, Baxter and Scott (1988a, the latter co-author is missing in the reference (also in the 1988b reference)) should be cited here. See also my general comment above, suggesting comparison of these measurements to the model simulations presented here.

The referenced citations are correctly included in the manuscript now and the general comment has been addressed above.

Page 30618, line 26: Mention "As" in the text.

Done.

Page 30619, line 4: Please note the revised half-life time (and thus decay rate)

Corrected. The revised half-life introduced an approximately 0.05% difference from before.

Section 2.2, first paragraph: Please explain abbreviations/acronyms of the model schemes (to provide at least some clue to non-modelers what they mean). It is also not clear how the nested grid of the model domain looks like.

We elaborated on the different schemes names and included additional figure to show our modeled domains (new Figure 1)

Section 2.2, second paragraph: The part starting in line 13, "This is partially . . ." is unclear.

We have rewritten the sentence.

Page 30621, lines 18-26: Taking into account the correct emission height of the nuclear facilities is of great importance: e.g. at 5 km distance from the source the ^{14}C excess may be up to one order of magnitude larger during stable situations for emission heights of 20m compared to 100m.

Indeed, emission height is of particular importance when looking at night-time / stable

situations, when the boundary layer height is limited and many large industrial emission stacks are built high enough to ensure that the emissions will be transported above, rather than stay in the stable boundary layer. This is true not only for the nuclear emissions, but for the fossil fuel emissions as well in this study, and in more general sense for all anthropogenic emissions. However, many modeling systems do not explicitly resolve the vertical distribution of emissions and some of the data is not even available in specific datasets. We now explicitly mention that we omit the vertical distribution in our methods and discussed later on its possible significance. We are working on a follow-up study to evaluate its importance when used in our system.

Page 30623, line 9: Guess the flux unit should be mg CO_2 *m-2 s-1.*

Indeed, this was an oversight. Thank you for mentioning it.

Page 30623, line 12: The fact that NEE is over-estimated during cloudy conditions may cause a serious bias in modeled plant material ^{14}C *as these weather conditions may be always associated with certain wind directions or catchments and thus particular source influence.*

Indeed, the covariations between weather, plant growth, and source signatures could be significant and bias results. In the study connected with our previous paper (Bozhinova et al., 2013), we explicitly tested the sensitivity of the resulting signatures to errors in the supplied weather data. In the described by the reviewer situation, the largest error introduced would be from prescribing larger daily radiation sum to the crop model, because the weather model does not always simulate the cloudy conditions accurately. In that case the crop model would simulate higher CO_2 assimilation, while in reality the plant growing conditions would not be as beneficial. In our previous sensitivity tests we found that while this will have large impact on the final biomass acquired by the plant, the impact on the modeled $\Delta^{14}CO_2$ was low, mostly due to the nature of the integrated signals stored in the plant.

Page 30624, lines 8-12: I am not convinced by this statement: The seasonal cycle at

Jungfraujoch is largely determined by the pre-set background values (as also stated by the authors) and at Schauinsland the agreement between model simulations and observations is not really good. Table 1 states a mismatch of appr. 2 ‰ which corresponds to more than 30% of the total signal (i.e. ^{14}C *deviation from background). Why does this indicate that the background location is representative for the region?*

Indeed the previous sentence was unclear. We based our statement on the applicability of Jungfraujoch data to serve as the signature of the background CO_2 fields based on the model results that do not show significant local or transported influence of anthropogenic (fossil fuel or nuclear) at the site. On the other hand, the average mismatch at Schauinsland indicates that the model overestimates the fossil fuel influence in the area. Because the mismatch remains close to the limits set by the instrumental uncertainty in the observations, we considered the model performance relatively good. Now that we have revised the paragraph to include new observational data, we also revised this sentence. It now reads: "The comparison shows we capture reasonably well the seasonal trend for most sites, however the model genrally underestimates $\Delta^{14}CO_2$. This might partly be caused by the omitted biospheric disequilibrium term, which accounts on average for up to 1.5 ‰ at these latitudes. Additional bias could be introduced through our choice of background site. . . ."

Page 30624, lines 12-29: I do not agree that the CO-based FFCO_2 *data shown in Figure 3b are independent from the* ^{14}C *data at Lutjewad (at least if these are the same data as published by van der Laan et al. (2010, Figure 1E)). The high-resolution CO-based data have been corrected/calibrated with* ^{14}C *observations and thus, on average, should give the same FF*CO_2 *concentrations as the monthly mean* ^{14}C *data directly. Indeed, it seems that also the "measurements" (blue dots) are considerably higher than the model results (red line), although no statistical information is listed in Table 1 for this comparison. Principally the same information as in Figure 2 is given in panel c of Fig. 3. So, indeed, the model underestimates FF*CO_2 *at Lutjewad, but note again that the CO-based estimates are not independent from the* ^{14}C *results (last line*

on Page 30624)!

The CO-based FFCO_2 data is indeed corrected/calibrated with ^{14}C observations from the same location. However, these observations were from sector-specific measurements (south wind sector measurements), rather than the continuous integrated samples presented here, as stated in the referenced van der Laan et al. (2010). As such we believe that our statement regarding the independence of the observations is true, as the information used to obtain the FFCO_2 data is not the same as the one it is compared to. The 6-month average daily statistics of the comparison between this data and our model are already included in Table 1, but among the CO_2 concentration comparisons. We have now corrected this by moving this data in a separate section in Table 1 to make it more obvious that the data is different than the rest of the CO_2 observations. Additionally, we looked at the observations-to-model comparison on a monthly basis (rather than for the whole period as the statistics in the Table 1) and at the start of the period the model performance is indeed worse, with mean monthly mismatches of -4 to -7 ppm (negative sign indicates underestimation in the model), but in the latter four months the monthly average mismatches are within -1.5 to -2.5 ppm difference. It is important to note, though, that this observational data contains considerable uncertainty as the observational data is calibrated with a 3-yearly fit of the ^{14}C-CO ratio, which on a bi-weekly scale can lead to up to three times overestimation of the fossil fuel CO_2 (as in April and start of May).

Page 30625, first paragraph: Again, I do not see good agreement in Figure 3b. Why should there be a problem with the ^{14}C observations themselves (from one of the best Radiocarbon Labs world-wide)? Please clarify and give a justification for this suspicion (may be one of the co-authors from Groningen can help here).

See our elaborate response above

Page 30626, line 2: Please give a bit more quantitative information about the size of the "area around the nuclear sources". This is critical for choosing the proper sites for

future ^{14}C monitoring.

For most pressurized and boiling water reactors we find that the nuclear influence is mostly contained within the grid cell of the source (12x12 km2), but will probably be confined in even smaller area if the horizontal resolution of the model is enhanced. For the gas-cooled reactors, which emissions are sometimes more than 10 times higher than the previously mentioned, we see that the influence can be expected even outside a 50 km radius. We have decided to add this additional explanation in the referenced paragraph. Additionally, we have included a new panel in Fig. 9, in which we show the difference in plant signatures modeled with and without including the nuclear emissions in the simulation.

Page 30627, lines 21-25: I am not sure that this explanation is correct. In fact, in Heidelberg we are sampling separately over day and night and find appr. 2‰ higher values in the daytime samples compared to nighttime samples during the summer half year (i.e. similar value as presented in Figure 7. By the way: please name the scale "$\Delta\Delta$" as it is the difference between plant and atmosphere). My explanation for the difference is that during day in summer the fossil fuel CO_2 signal is much more diluted than during night. The authors may test this hypothesis/alternative explanation by simply comparing daytime and whole model results.

We realize that by stating "the atmospheric mean $\Delta^{14}CO_2$ for the growing period of the plant" in this paragraph we weren't clear enough. Here we use the atmospheric average for the period when the plant was growing, which includes the daytime period only for the length of the growing period simulated by our crop model. This is connected with another specific comment regarding the conclusions. We have revised the paragraph and the specified conclusion to make this message clearer. We have also corrected the figure colorbar label as suggested. Nevertheless, we should note that indeed, there is difference on the average between daytime and nighttime $\Delta^{14}CO_2$ signatures, which is also found in our model. The abovementioned figure, however, does not deal with this issue as it compares directly modeled daytime atmosphere to plant signatures.

Page 30628, Section 3.4: I am not sure that this section is really needed/useful. My feeling is that it may be rather counter-productive because it supports (although it does not intend to) the frequent misconception that boundary layer concentrations can be simply set equal/proportional to ground level fluxes.

Indeed, we understand 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 present the results of comparing a simple box model to the results from the more comprehensive transport-modeling framework.

Page 30630, lines 13-15: It should be emphasized here that it is also very important to release the $^{14}CO_2$ from nuclear facilities at the correct height level (see earlier comment).

The paragraph is revised to include more information on the vertical spatial and temporal resolution of the data and now states explicitly what is not accounted for in our study.

Page 30632, line 25ff: Why do we need flask samples to find out from which direction nuclear enrichment could be? I guess we know very well the location of all our large point sources (also for fossil CO_2 emissions).

While the sources associated with nuclear industry are well known, there are other sources of anthropogenic $^{14}CO_2$, which even though considerably lower on the global scale, can influence local observations – such as experimental reactors, used for research purposes in universities and research institutes, or medical facilities that produce/use radioactive particles for operational use.

Page 30633, line 17-21: I am not so sure that the difference between plant and integrated atmospheric samples is not mainly due to the diurnal cycle of atmospheric

mixing (see comment above).

While your statement is certainly true, the referenced lines concern a conclusion in which we compare the differences between the modeled plant sample and the modeled atmospheric average for the period the plant sample was assimilating CO_2. This thus also includes only daytime data and also considers the main period in which the plant assimilated CO_2. We realize this was not communicated well in the presentation of the results and the conclusion and we have revised the connected paragraphs.

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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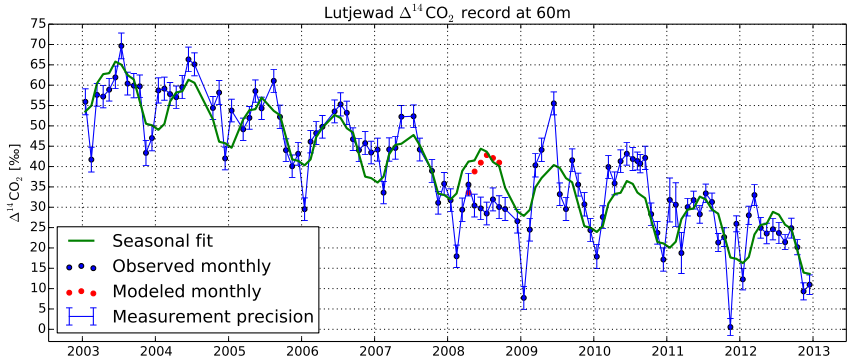

Fig. 1. Lutjewad Δ14CO2 observations and functional fit, in addition to modeled values for 2008 (source of observational data: Sanne Palstra, Centre for Isotope Research, Groningen, personal communicat

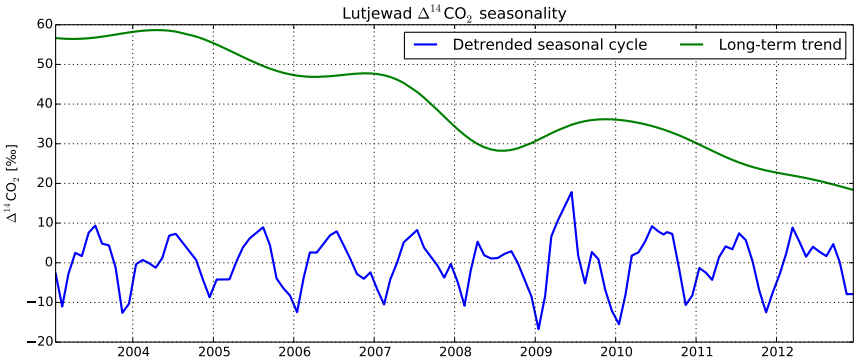

Fig. 2. De-trended seasonal cycle and long-term trend of the Lutjewad Δ14CO2 observations