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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. Vogel for his constructive assessment of our work and the insightful comments that allowed us to improve our manuscript. We address his questions in detail below.

General comments: 1. This study should address the limitations of using only 6 month of (summer) data in greater detail. The fossil fuel CO_2 *emissions, especially in large urban areas increase substantially during winter (cold) month. The local and large-scale CO2ff gradients can be expected to be significantly different then. The different atmospheric conditions (e.g. more synoptic, rather than daily variations of trace gas concentrations) can also alter the results of a model-data comparison. Referencing*

e.g. "summer" in the title and a short discussion could help clarify this.

As our intention is to use the model results to simulate also plant samples, the growing season of the crops chosen for our study was a determining factor for the period of the simulation. We agree that in the study of anthropogenic emissions the wintertime is also an interesting period as both the signals from anthropogenic origin are stronger, and those from biospheric sources are smaller, and without the suggested change in the title it could be misleading to potential readers. We have thus revised our title to include 'summertime'.

2. Given the high spatial and temporal resolution of the modelling framework the limitations of using a parameterized emission estimate of nuclear power plant $^{14}CO_2$*, can cause several problems (Vogel et al. 2013, Radiocarbon) This study, unfortunately, does not account for the large uncertainty of the parameterization as reported by Graven and Gruber (2011) and thus likely underestimates the uncertainty this term contributes to the uncertainty of D14C. This study, furthermore, assumes a constant annual emission of* $^{14}CO_2$*. Although, the* ^{14}C *is produced relatively constant over time, its release can often be intermittent and linked to maintenance work e.g. Vogel et al. and references therein. If no higher resolution emission data can be retrieved it seems crucial to use the uncertainty range provided by Graven and Gruber (2011) to estimate the uncertainty of the d14Cnuclear term.*

While we discuss the problems associated with the unknown temporal variability of the nuclear data in length in our Discussion, it was an omission not to include the sensitivity of our results to the uncertainty in the reported emission factors. We have now included two additional nuclear estimates (for the low and high end of the 70% confidence interval associated with the emission factors used) and re-ran our simulation for the last two months of the period – when the winds are mostly westerly and the nuclear influence over the domain is considerable. The results are now shown in a new Figure 6 and explained in the additional paragraph in Section 3.2. While this analysis has revealed the higher uncertainty of the nuclear estimates in Southern Germany, Central France

and especially in the United Kingdom, our biggest source of nuclear emissions in the domain – the fuel reprocessing site of La Hague – is reporting directly ^{14}C emissions and thus, not subject to the uncertainty in the emission factors. We have to add that in over 95% of the domain the differences between both high/low and the control run did not exceed 2 ‰ and within 99.5% of the domain it did not exceed 5 ‰

3. The methodology presented in Section 3.4, which derives CO2ff fluxes from mere concentrations could be misleading. Given that this section uses only synthetic data finding a good relationship between concentration and fluxes, when accounting for the "footprint" (=5x5 pixel average) seems straightforward. The fit should even be better when the real footprint is used for calculating the flux average instead the 5x5pixel mask. The concentration is afterall the convolution of footprint and flux. It is, however, unclear to me how this method could improve our understanding of fluxes, as deriving fluxes from concentrations does usually not work in the real world, but often requires more complex models than a linear fit.

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.

Overall the modelling framework presented here has a great potential to help better understand atmospheric D14C and the ability to use D14C data to derive fossil fuel CO_2 fluxes. Comparing different potential sampling strategies and techniques will be crucial to develop monitoring schemes to quantify fossil fuel CO_2 emissions from atmospheric observations. After addressing the general comments this study will most definitely be a valuable addition to this field and a suitable contribution to ACP. Specific comments: Title: Please consider adding a reference to the limited time of the simulation here.

Our title is now “Simulating the integrated summertime $\Delta^{14}CO_2$ signature from anthropogenic emissions over Western Europe. We have also acknowledged the limited simulation in our Methods and also included it in the Discussion as follows “However, the use of plant samples is typically limited to the summertime, which is a period with lower anthropogenic CO_2 emissions, more vertical mixing, and larger biospheric fluxes. This will correspond to larger uncertainty in the recalculation of the fossil fuel CO_2 emissions compared to wintertime.”

P30613 - line 2-5 Giving an estimate of the typical uncertainty at this scale would help the reader to appreciate which precision the top-down method has to achieve to be useful to improve the bottom-up estimates.

Indeed, an estimate from one of the cited papers is now included in the sentence.

P30614 - line 1-4 Please add "and upper troposphere", as a notable amount of ^{14}C is produced there es well. Tropospheric $^{14}CO_2$ also tends to be transported to lower levels more quickly.

Corrected.

P30619 - line 25 Please add the information about the spatial resolution of the used meteorological data here.

The resolution of the NCEP data is now included in the text.

P30620 line 24-26 The biospheric flux model uses different meteorological data than the atmospheric transport model. Do you have an estimate of the differences of important variables (T, wind, solar radiation, ...) of those two meteorological datasets?

We do not have a comparison available of the different meteorological datasets over our domain, although of course much work is focused on comparing weather forecasts. We note though that the meteorology driving the boundary conditions of the atmosphere (NCEP) and the monthly mean biosphere model fluxes (ECMWF) are both based on reanalysis and should include many observations from our domain to stay close to

observed weather. The WRF model with its own high-resolution forecast presumably stays close to this reanalysis because the synoptic scale events (high+low pressure systems, fronts) are applied through the boundary conditions. For the biosphere, the high-resolution WRF meteorology is moreover only used to determine the hour-to-hour variability and not the mean fluxes (these come from SIBCASA).

P30621 - line 18-26 See general comment 2. The production of ^{14}C might be continuous, the release likely has both constant and large intermittent components. (Vogel et al. 2013, Radiocarbon, and references therein)

We are aware of these differences and have discussed them in a following section. For clarity, we now include a more explicit overview in the methods.

P30624 - line 10 and Figure 2.d The information that Jungfraujoch is used as Dbg is substantial. Please consider removing the parenthesis. Given the small addition of co2ff and 14co2nuc at JFJ, an agreement of the data from JFJ and a model driven by Dbg from JFJ is rather to be expected.

Corrected.

P30625 - line24 following and Figure 2 and Table 1. The mismatch of D14C is quite large and an explanation seems hard. The Van der Laan et al. 2010 (VDL10) data seems to agree significantly better, although its seasonality is also determined by a ^{14}C calibration. The model has a bias of 8.82permil to the ^{14}C data from Lutjewad presented here. The mean bias to the hourly VDL10 data seems to be fairly consistent i.e. -2.31ppm CO2ff, which translates to roughly 6permil- 7permil. Do you have a comparison of the D14C data used to calibrated VDL10 and the samples used in this study?

We would like first to clarify the statistics of the comparison with the data from the van der Laan et al. (2010) study. The statistics shown in our Table 1 are the statistics of the daily mean values observed and modeled for the entire period of 6 months (or

slightly shorter because in some cases observational data was missing). When we did the statistical comparison month per month, we noticed that the first two months were showing much higher mean bias -4.5 to -7 ppm (the negative sign indicates that the model underestimated the quantity compared to observations) of fossil fuel CO_2, than later months when it was on average between -1.5 to -2.5 ppm. This is probably connected with the ^{14}C-CO calibration of the observational data – as described in van der Laan et al. (2010), the resulting fossil fuel CO_2 observations are obtained using 3-yearly fit to the observed bi-weekly ^{14}C-CO ratio. In some cases, as is for late April and early May, this will result in the fit being three times lower than the actual observed ratio and thus resulting in overestimation of the fossil fuel CO_2. However, even if we use the overestimated fossil fuel CO_2 to calculate the amount missing between the model and observations, it could account for only half the difference in the mismatch in $\Delta^{14}CO_2$ in the later months, while in the earlier months the mismatch in $\Delta^{14}CO_2$ is lower.

We investigated the issue with the model-to-observations mismatch for the station of Lutjewad in our simulation of summertime 2008 $\Delta^{14}CO_2$ signatures. Our colleagues 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 obtain the functional fit to the data and additionally, the de-trended seasonal cycle (shown in the following to figures). 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 similar no-peak year occurs in 2012 as well, so it is unlikely it is something connected with the observations themselves. Our Groningen co-authors have confirmed that in principle they see no measurement related problems with the data presented, but they also do not have a physical explanation for the

observed feature 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 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.

Section 3.4. See general comments 3.

See the replies to the general comments.

P30630 - line 15-16 Please add a reference. According to (Pregger and Friedrich 2009, Environmental Pollution, doi:10.1016/j.envpol.2008.09.027) and others, a significant amount of emissions are emitted above 300m.

Indeed, the numbers cited in this part are a direct estimate from the database used and are average heights at the spatial resolution provided. This clarification is now included in the paragraph.

P30630 line 22-24 See general comment 2. Please reconsider the "safe assumption" of constant $^{14}CO_2$ emission from nuclear power plants or add citations of respective literature to back this assumption.

We have elaborated and revised this paragraph.

P30631 - line 2-3 Please address that using plants for monitoring $^{14}CO_2$ will be limited to summer month (in Europe) and can thus only be complementary to other techniques.

Indeed, this is always the case. The advantage of having plant samples in addition

to your regular sampling techniques is that it can provide some spatial resolution in a very scarce observational network. Unfortunately, the disadvantage is that this can also happen only during a certain period throughout the year, and particularly not the period when the fossil emissions are the most significant contributor in the land-atmosphere CO_2 exchange (winter).

Figure 3 - caption Please change "contrubution" to "contribution"

Corrected.

Figures general Please add a, b, c to subfigures to identify them

The letters that identify each panel, as used in the captions, are now included in the panel's title.

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 30611, 2013.

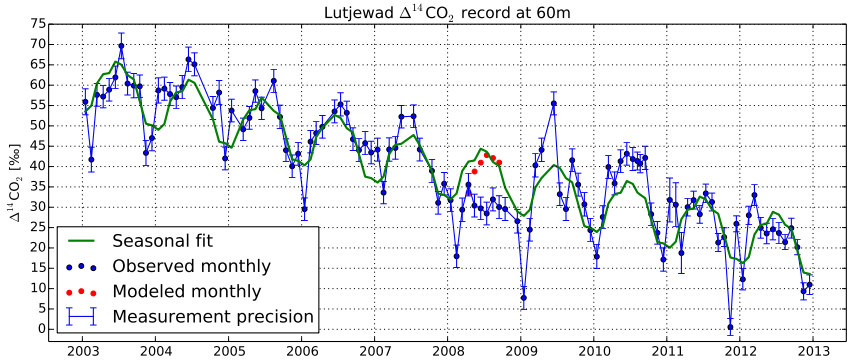

Fig. 1. Lutjewad Δ14CO2 observations and functional fit, in addition to modeled values for 2008 (source of observational data: Sanne Palstra, Centre for Isotope Research, Groningen, personal communication)

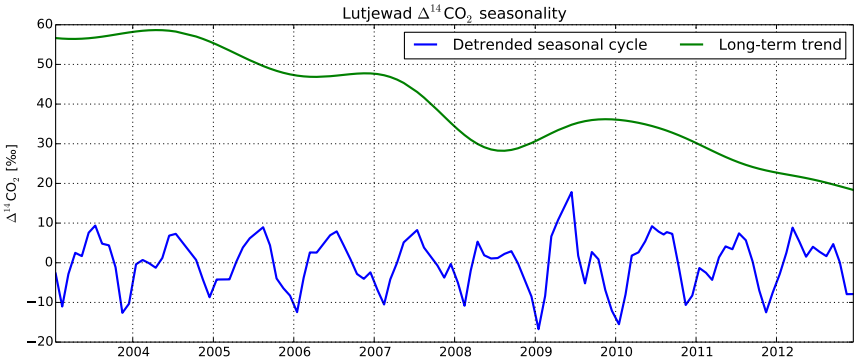

Fig. 2. De-trended seasonal cycle and long-term trend of the Lutjewad Δ14CO2 observations