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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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This paper describes a regional (WRF-CHEM) modeling study which simulates 14CO2 and CO2 over Europe. They use the model results to consider: (a) optimal sampling methods for 14CO2 (flasks vs integrated vs plant samples); (b) how important nuclear industry 14C production is in the European region. They test a novel idea for comparing atmospheric 14CO2 observations with reported fossil fuel CO2 emissions, looking at the (simulated) relationship between reported emissions and atmospheric 14CO2 measurements across a range of different European urban areas. Finally, they use the model to suggest overall optimal sampling strategies for 14CO2 in the context of constraining fossil fuel CO2 emissions.

This paper is certainly worthy of publication in ACP. 14CO2 modeling studies are still few and far between, and this is one of only a handful of regional 14CO2 studies. The detailed analysis of the significant European nuclear industry influence on 14CO2 is sorely needed.

General comments: Clearly, nuclear industry emissions are an important influence on Δ14CO2 in Europe, particularly for Britain and France. However, when fossil fuel CO2 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 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.

Section 3.4. Direct estimation of fossil fuel CO2 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 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.

Specific comments. Pg 30613, lines 14-25. The phrasing of this section seems to imply that correlate tracers are better than 14C as fossil fuel CO2 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 14C. It is also worth noting that only some of these tracers are co-emitted with fossil fuel CO2, with others having only approximately co-located, but independent, sources.

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.

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

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.

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

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.

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 14C 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 (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)?

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 14C 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.

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.

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

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

Pg 30630, lines 22-24. I'm not sure that it is true that reactor 14C 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.

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 CO2. 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?

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 14C measurements is also needed – see my general comment.

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.

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