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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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The manuscript presents a comprehensive modeling study of the combined effect of fossil fuel CO2 emissions (that lack 14C) and 14CO2 emissions from nuclear installations on the boundary layer 14CO2 concentration over Western Europe. It aims at supporting the interpretation of 14C observations in atmospheric CO2 and plant material, collected for estimating the amount of regional fossil fuel CO2 in the boundary layer atmosphere. The authors use a spatially and temporally adequately resolved modeling framework of atmospheric transport, combined with a recently developed crop growth model as well as high-resolution European emission inventories for fossil fuel CO2 as well as 14C emission estimates from nuclear installations.

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 14C 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 14C 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. Unfortunately, the comparison with 14C data and CO-based fossil fuel CO2 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.

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).

Page 30613, lines 18-20: I do not agree that it is an advantage that e.g. the CO/FFCO2 ratio for traffic emissions is so much different from that of other FFCO2 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/FFCO2 ratio with 14C data on the seasonal and diurnal time scale to account for the temporally changing source mix.

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

Page 30614, line 3: Only slightly more than 50% of 14C 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).

Page 30614, lines 17-18: Please note that (at least to my knowledge) there is no 14C counting technique in use that is measuring graphite targets.

Page 30615, line 3: Here a CO2 term from the stratosphere is missing.

Page 30615, line 23ff: It is not totally clear if all components of the ecosystem respiration 14CO2 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.

Page 30618, lines 1-3: It should be mentioned here, that e.g. for the Heidelberg fossil fuel CO2 estimates – i. e. already in Levin et al. (2003) - we made a correction based on measured (!) monthly 14CO2 emission data from the NPP Philippsburg and a Gaussian Plume model.

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.

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

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

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.

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

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 14C excess may be up to one order of magnitude larger during stablesituations for emission heights of 20m compared to 100m.

Page 30623, line 9: Guess the flux unit should be mg CO2 m-2 s-1.

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

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. 14C deviation from background). Why does this indicate that the background location is representative for the region?

Page 30624, lines 12-29: I do not agree that the CO-based FFCO2 data shown in Figure 3b are independent from the 14C 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 14C observations and thus, on average, should give the same FFCO2 concentrations as the monthly mean 14C 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 FFCO2 at Lutjewad, but note again that the CO-based estimates are not independent from the 14C results (last line on Page 30624)!

Page 30625, first paragraph: Again, I do not see good agreement in Figure 3b. Why should there be a problem with the 14C 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).

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 14C monitoring.

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 CO2 signal is much more diluted than during night. The authors may test this hypothesis/alternative explanation by simply comparing daytime and whole model results.

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 mis-conception that boundary layer concentrations can be simply set equal/proportional to ground level fluxes.

Page 30630, lines 13-15: It should be emphasized here that it is also very important to release the 14CO2 from nuclear facilities at the correct height level (see earlier comment).

Page 30630, 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 CO2 emissions).

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).

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