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Interactive comment on “Source apportionment of elevated wintertime PAHs by compound-specific radiocarbon analysis” by R. J. Sheesley et al.

Anonymous Referee #1

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General comments:

The manuscript "Source apportionment of elevated wintertime PAHs by compound-specific radiocarbon analysis" by Sheesley et al. describes sampling of particulate matter at the rural site Lycksele, Sweden during winter and the isolation of TOC and different PAHs for ^{14}C measurement. The latter is done by multiple injections of extracts into a PCGC instrument, evaporation of solvents and accelerator mass spectrometry analysis. The following PAHs were analyzed for their biomass-burning contribution: phenanthrene (gas phase), phenanthrene (aerosol phase), fluoranthene (aerosol phase), pyrene (aerosol phase), fluoranthene (aerosol phase), benzo[b+k]fluoranthene composite (aerosol phase), and indeno[cd]pyrene + benzo[ghi]perylene composite (aerosol phase). In general, PAHs showed a strong non-fossil signal with 71%-87%

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biomass-burning contribution due to the dominant emissions of particulate matter from residential wood combustion. Although the sample number is very low, the measurements are so unique (to my knowledge, ^{14}C analyses of individual PAHs have not yet been performed on ambient particulate matter filters) that publication of the study is justified. However, some weak points have to be improved before the manuscript can be accepted.

Specific comments:

1. P20902L18-19: This sentence is misleading, as one may understand that this fraction has a contribution of biomass combustion of 9%. It should be clarified like: "9% lower contribution of biomass combustion than the sum of all PAHs".
2. P20902L21-23: This sentence should be removed as it does not contain any clear message.
3. P20907L28-P20908L3: The authors should also present results of the blank contribution (carbon mass and isotopic value) for the PCGC procedure. It is clear that 40 injections per extract introduce contamination into the sample. Only if these numbers are given, one can evaluate whether this contribution is significant.
4. P20908L15-17, P20912L25: Substitute "error" with "uncertainty"
5. P20909L24-27, Figure S1: Several studies are cited, which use CMB modeling to estimate the contribution of biomass burning. As the dataset of this work includes concentration determinations of different PAHs, the authors should provide such a CMB modeling estimation from their data (e.g. by using the PAH/OC ratios of Figure S1 or by following the concept of Schauer and coworkers) as an alternative approach in order to compare it with the ^{14}C results.
6. P20911L15: Remove the word "then".
7. P20911L21: The reference value for $\Delta^{14}\text{C}_{\text{fossil}}$ should be repeated here as well.

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8. P20912: I recommend compiling all data in a table (considering SRM1649a, Kumata et al. (2006), all papers of Reddy et al., all papers of Mandelakis et al., etc.). It is important to note that the older data of SRM1649a (e.g. in Currie et al., 1999) were biased by an erroneous blank correction so that results of Currie et al., J. Res. Natl. Inst. Stand. Technol. 107, 279-298, 2002 should be used. The compilation will help to distinguish CSRA from CCSRA results and to show the broad range of ^{14}C data for the PAHs. Especially, the totally different levels for SRM1649a (95% fossil) and samples at Lycksele (20% fossil) should be addressed.

9. P20912L11: Does "35-65%" refer to TOC or PAH?

10. P20912L15: The reference of the word "respectively" is unclear.

11. P20912L22: The sentence should begin like this: "To evaluate the isotopic..."

12. P20913L20-25, Figure S1, P20902L16-18: I assume that there are more studies on PAH/OC ratios of diesel and gasoline exhaust emissions than Lough et al. (2007). Results of other publications should be included in Figure S1, which should be considered in the CMB estimations and in the diesel vs. gasoline discussion. It is a weak point that Lough et al. (2007) do not present results for B[ghi]P, as I can achieve from Figure S1. As this PAH is analyzed together with I[cd]P, any interpretation of the ^{14}C data of this PAH combination is only valid, if emission ratios for both PAHs can be found. Due to this, the corresponding clause in the abstract ("known to be enhanced in gasoline-powered motor vehicle exhaust compared to diesel exhaust") should be removed, unless better data are presented.

13. P20913L26-P20914L2: This sentence should be removed as it contains too much speculation for the given ^{14}C results of this study. To me, car fleet numbers will provide more reliable and statistically significant results than the PAH/OC ratios in combination with the ^{14}C values of the B[ghi]P+I[cd]P combination.

14. P20914L21-23: The power of this statement contradicts the interpretation of the

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data in P20913L7-8 ("...inhibits statistical analysis"). The sentence should be weakened.

15. P20915L5: The power of this statement contradicts the interpretation of the data as already discussed in items 12 and 13. The sentence should be weakened.

16. P20915L8-11: New discussion threads as well as literature citations are not acceptable in the Conclusions.

17. Table 1 and section 2.4: Considering all the numbers given in the manuscript, I estimate that 30 μgC were used for the ^{14}C analyses of the individual PAHs. If this is true, it is disappointing due to the fact that there are several AMS laboratories in the world performing ^{14}C measurements for samples $<10 \mu\text{gC}$ on routine basis. This means that measurements could have been performed on the fortnight filters! Sample masses should be given in Table 1 or in the supplementary material.

18. Table 2: New data from the study should be marked more clearly, e.g. by "this work".

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