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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source Apportionment of Carbonaceous Aerosols in Beijing with Radiocarbon and Organic Tracers: Insight into the Differences between Urban and Rural Sites" *by* Siqi Hou et al.

Anonymous Referee #2

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The authors incorporated the 14C analysis into the source apportionment method developed by Gelencsér et al. to apportion the fossil and non-fossil sources of EC and primary and secondary OC in Beijing. A total of 25 PM2.5 samples collected at an urban and a rural site in summer and winter were analyzed. An evident seasonal variation of fossil/non-fossil and primary/secondary OC was observed for both sites. Although the method used in this work is at the preliminary stage, it provides a new way to apportion primary and secondary OC from fossil and non-fossil sources. The relatively good agreements between the source apportionment results obtained from this

work and CMB models suggest the feasibility of this 14C analysis-incorporated Gelencsér method. The current model only includes levoglucosan as the source marker for biomass burning. Inclusion of more source markers in this method, such as hopanes for gasoline vehicles, cholesterol for cooking, and Al and Si for soil dust, may give a more explicit source apportionment of non-fossil and fossil-derived OC. Overall, the manuscript is well written, and I recommended it for publication with some comments provided below.

Major comments:

1. Previous studies on the source apportionment of PM2.5 in China showed that dust (including soil dust and road dust) is an important contributor to carbonaceous aerosols in Northern China (Huang et al., 2014; Zhang and Cao, 2015). Its contributions to both PM2.5 and organic matter in Beijing were even higher than that from the cooking source when PM2.5 mass concentrations ranged from 60 to 200 ug m-3 (Huang et al., 2014). So why contributions of dust to EC and OC were not counted in this study? One of the major assumptions made in this study is that all ECnf is from ECbb, which ignored the contribution from dust and may overestimate ECbb. Since OCbb was derived from ECbb, and the difference between POCnf and OCbb was 100% attributed to OCck, which overestimated the contributions from the cooking source. This is reflected by the poor correlation between OCck-this study and OCck-CMB. As shown in figure 5e, most of the OCck values derived in this study were larger than those apportioned by CMB, which further proved that other sources, e.g., dust, that contributed to OCnf and ECnf have been overlooked in this method. Si and Al are good markers for soil dust. Similar to what the authors did for OCbb, the authors may try to use the measured concentrations of Al and Si, and the ratios of these two markers to OC or EC in the dust source profiles to derived OCdust and ECdust. Similarly, cholesterol (data reported in Wu et al., 2020) and its ratios to OC and EC in cooking emission profiles can be used to derive OCcooking and ECcooking.

Zhang, Y.L. and Cao, F., Fine particulate matter (PM2.5) in China at a city level, Sci.

Rep., 5, 14884, 2015.

Huang et al., 2014 and Wu et al., 2020, see the references in the manuscript.

2. Some of the key parameters, such as the fractions of levoglucosan from softwood burning and straw burning, were empirically derived using these 25 samples; emission source profiles of softwood burning, straw burning, and maize burning were also empirically selected based on their best fit to the measured ECnf values of these 25 samples. These parameters and source profiles were then used to apportion the non-fossil and fossil-derived POC, SOC, and EC from the same batch of 25 PM2.5 samples. If possible, another set of Beijing PM2.5 samples could be analyzed using the parameters and source profiles determined in this study to validate the extended Gelencsér method and prove its general application to Beijing samples.

Minor comments:

1. Line 143-144: How much filter was extracted by water? By which extraction technique and for how long?

2. Line 147-148, How great will these factors influence the accuracy of the results? Which one is most crucial?

3. The authors mentioned several times (e.g., line 70 and 249) in the manuscript that coal combustion must be included in the extended Gelencsér method. However, in this study, coal combustion still cannot be explicitly differentiated from the fossil-derived OC and EC from vehicle emissions. How would the author improve this?

3. Since the focus is on the 25 selected PM2.5 samples in this study, it makes more sense to me to present the compound concentrations and meteorological parameters of just these 25 samples other than the whole batch of samples in Table 2.

4. The authors may think about moving Table 3 to the SI, since it is not critical for the discussion. 5. I suggest the authors include the time series of apportioned POCf, SOCf, OCbb, OCck, and SOCnf at IAP and PG in figure 6 as well. This will help the

readers to follow the discussion more easily.

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