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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Fossil and Non-fossil Sources of Organic and Elemental Carbon Aerosols in Beijing, Shanghai and Guangzhou: Seasonal Variation of Carbon Source" *by* Di Liu et al.

Anonymous Referee #1

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General comments

In this work the authors collected 24-hr, PM2.5 samples in three urban centers for one month each season from October 2013 to July 2014. All samples were weighed for mass and analyzed using thermal-optical transmission for organic carbon (OC) and elemental carbon (EC). Two samples "with relatively higher and lower PM2.5 concentrations" in each season in each city were then selected for analysis of the water soluble (WSOC) and insoluble (WIOC) organic carbon content, and these fractions were further analyzed for the radiocarbon (14C) content. Radiocarbon allows for the separation

of the contributions from fossil carbon sources, e.g., coal, and biogenic carbon sources, e.g., biomass burning.

The source apportionment of carbonaceous PM2.5 is an important and very challenging task. The division of the PM2.5 carbon aerosol samples into the various fractions, i.e., EC, WSOC, etc., combined with the estimation of the 14C content of each fraction, is a powerful method for assessing the contributions of major source types to the PM2.5 carbon aerosols. The authors used sound methods for filter analysis that have been used in previously pubished studies. It appears that a subset of these data or similar data have been previously published by the authors including Liu et al. (2014), EST; Liu et al. (2017a), ACP; Liu et al. (2016b) EST; and Zhang et al. (2015a), ACP, which diminishes the scientific contribution of this work. One important contribution is the examination of the variations in these carbon fractions and 14C content across the four seasons and three urban centers. However, only two 24-hr samples were analyzed in each season for each urban center. This is not enough data to assess whether the differences are due to day to day variations or represent true seasonal and urban differences. Therefore, conclusions such as "Above all, this study demonstrates that the main sources of carbonaceous aerosol in cities varied greatly across different seasons, but the carbon sources of haze and non-haze days in each season showed little difference" (see lines 266–268) are questionable.

Specific Comments

-Due to the number of previous publications by the authors on similar data collected at the same urban centers from 2012 to 2014, I think the introduction should contain a brief review of these results and the unique contribution of this. A table listing exactly what days were analyzed at each urban center in each publication would be valuable and aid the discussion on page 10.

-Two 24-hr samples are not enough data to generate robust seasonal averages. It would be good for the authors to explore the possibility of aggregating results from this

and previous studies to generate more-meaningful seasonal averages and justifiable results.

-The filter analysis methods, such as TOT, are not without issues and biases. A short discussion of the limitations and biases in all of the filter analysis methods used would aid the reader's interpretation of results.

-Line 101. Please provide some indication as to the importance of the OC on the filter blanks. For example, what was the fraction of OC on the blank compared to the typical filter loading on an exposed filter? Were the filter blanks analyzed for WSOC and WIOC and the 14C fractions? If so, what were these levels relative to the exposed filters and were these data used to blank-correct the 14C data?

-Lines 136–137. "total organic matter (TOM = 1.6X OC + EC)". Note that EC is theoretically not part of TOM. I suggest this be renamed to something along the lines of total carbonaceous matter.

-Lines 136–190. In general the manuscript was easy to understand. However, this section was poorly written, making it very difficult to understand and review. Some issues include the introduction of the terms "haze" and "haze formation" with no definition and the mixing of TOM/PM2.5 and TC/PM2.5 ratios. Is haze synonymous with PM2.5?

-Lines 144–148. "Whereas in Guangzhou, the ratio of TC/PM2.5 was positively correlated with PM2.5 concentration ($R2 = 0.27$, $p < 0.05$). This means that relative contributions of carbonaceous aerosols to total fine particles increased when the haze occurred in Guangzhou, implying the role of carbonaceous aerosols is more important in South China than those in other parts of China." It would seem that it is the TOM/PM2.5 ratio that defines the importance of TOM to PM2.5 (haze), not the correlation of TOM/PM2.5 to PM2.5.

-Lines 185–190. The conclusions drawn from Fig. 1 are not obvious and deserve some additional discussion. In addition, due to the limited number of days used in

this analysis, statements such as "It means that the major sources of carbonaceous aerosol in autumn and winter came from inland China and from the Pearl River Delta in spring and summer" are not robust. This analysis could be done using all samples collected in each season, which would result in more-defensible conclusions.

-Lines 215–218. As mentioned before, I question statements such as these based on the limited number of samples used in the analysis.

-Line 264. "However, the carbon sources during haze and non-haze..." Haze and non-haze were never defined.

Technical Corrections

Line 60. "disintegrated" is not the proper term. Maybe use "decomposed" instead

Line 162. Should "Adversely" be "Conversely"?

Lines 165–67. Repetitious sentences.

Line 293. What is meant by "incensement"?

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