

In this study, different fractions of organic carbon in ambient fine particulate matter (PM<sub>2.5</sub>) in ten Chinese cities and their optical effects were investigated. Results indicated that the optical effects of extractable organic carbon are mainly contributed by relatively hydrophobic fractions (i.e., water-insoluble organic carbon and humic-like substances). Both empirical indices and the source apportionment model indicate that aromatic compounds from primary emissions tend to exhibit a stronger light-absorbing capacity. This study can provide significant information on the chemical compositions and sources of brown carbon (BrC) for further mitigating the climate effects of PM<sub>2.5</sub>. I recommend accepting this manuscript if the following comments could be addressed in the revised version.

General comments:

1. This study cannot represent the entire China, because only samples from urban areas were analyzed. All these samples were collected during 2013-2014, a period marked by intensive coal combustion in China. Since 2017, coal has been gradually replaced by natural gas for domestic heating during the cold season. Please change the title of the manuscript.
2. This manuscript only investigated the optical effects of organic aerosols. Please remove sentences regarding the health effects of organic aerosols from the Abstract and Introduction.
3. In this manuscript, coal combustion has been proposed as an important source. Please demonstrate the contribution of coal combustion areas without central heating during winter. Is coal combustion indeed a significant source?
4. Cl<sup>-</sup> is used as a marker of coal combustion. However, sea salt is also a significant source of Cl<sup>-</sup> in PM<sub>2.5</sub>, particularly in cities like Shanghai and Guangzhou. Please ensure that only non-sea salt Cl<sup>-</sup> is included into the model. Please refer to the equation for calculating non-sea salt Cl<sup>-</sup> as provided in Ma et al. (10.5194/acp-18-5607-2018).
5. The authors proposed a light-absorbing carbonaceous continuum. However, it is important to note that there may be overlaps between the different carbon components. The operational definition of carbon components also varies. The compounds the authors refer to as 'char BC' most likely belong to 'brown carbon' or 'humic like substances', and are unlikely to bias optically based BC

measurements in large cities. Please address this point.

6. Please double check for grammar. There are lots of grammatic errors in this manuscript.
7. Please shorten the titles of each section in Results and Discussion.

Specific comments:

Section of Materials and Methods:

1. Please clarify the year in which the samples were collected.
2. Why does Figure 1 display the average aerosol optical depth at 550 nm instead of other wavelengths?
3. Please describe the instrumental method of ion chromatography.
4. The random errors and rotational ambiguity of the source apportionment model should be estimated using the bootstrap model and the displacement model. Please provide the evaluation results.

Section of Results and Discussion

5. Please clarify whether the value after average is standard deviation or interquartile range. For instance, in Line 232-232, “The concentrations of WIOC ranged from 1.45 to 12.95  $\mu\text{gC}/\text{m}^3$ , with an average of  $3.64 \pm 2.53 \mu\text{gC}/\text{m}^3$  among the 10 cities (Figure 2a).”
6. BB in Line 279 and MW in Line 320 should be defined.
7.  $p$  should be in italic.

Technical corrections:

Line 20: Water-insoluble “organic” carbon

Line 45: WIOC “is” primarily originated

Line 157: the relative standard deviation of what?

Line 251: were higher than those of non-HULIS (Figure “2”c).

Line 426: It is important