

Interactive comment on “Characteristics and the origins of the carbonaceous aerosol at a rural site of PRD in summer 2006” by W. W. Hu et al.

Anonymous Referee #2

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General Comments

This manuscript presents semi-continuous measurements of carbonaceous aerosol components in a background site in the Pearl River Delta region of China in July 2006. They discuss the diurnal and daily variations in aerosol composition as a function of aerosol sources and meteorology. A modified EC-tracer method is used for estimating primary and secondary OC. The modified EC-tracer method appears to have significant drawbacks that include 1) an unsubstantiated hypothesis that primary combustion sources are not related to SOA formation and 2) omission of non-combustion sources of OC, which appear to be significant based on their data. Further, there is question as to the validity of the measurements made, as the authors present an unreasonable ratio of organic matter (OM) to organic carbon (OC). Their data indicates $OM = OC$,

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leaving no room for oxygen or hydrogen in OC, a result that is inconsistent with prior studies and contradictory to their conclusion that OC is oxygenated. The manuscript contains many grammatical errors that detract from its readability. Major revision is needed prior to publication.

Specific Comments

1. The manuscript requires significant revision with respect to grammar.
2. Figure 2, what is the cause of the breaks in the data (especially the measurements of PM 2.5, ECOC, and WSOC that are relevant to this manuscript)? How do these breaks affect the representativeness of the sample size?
3. Section 3.1- The 1.01 slope of the OM/OC correlation suggests that $OM=OC$, which means that OM is comprised of carbon alone. By definition, $OM > OC$, because OM also includes elements such as oxygen, hydrogen, and nitrogen. OM/OC ratios have been documented in the literature in the range of 1.2 to 2.0 (Bae et al., 2004; Turpin and Lim, 2001). The low end of the range represents emissions with strong primary influence with the high range suggestive of oxygenated or secondary sources. The data shown here is not consistent with the conclusions of the manuscript, such as SOC averaged 47% of OC, which would necessitate $OM/OC > 1.2$. Even having included high primary emission data, this result is unreasonable. The authors must explain this unexpectedly low OM/OC ratio and discuss the error in measurement.
4. What appears to be an error in AMS measurements of OM (discussed above) should also be discussed in the context of the other AMS measurements reported in this manuscript.
5. For the purpose of validating chemical measurements, the authors should perform a mass balance on PM_{2.5}, in which they reconstruct PM_{2.5} mass from the measured components and discuss the difference as a function of unmeasured species and measurement uncertainty.

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6. Section 3.2- Designation of three types of days- How were the “source influences” determined?
7. Figure 3b, it would help the viewer to visualize the data if the x and y axes were set to the same maximum value (i.e. 60ug/m³) so that the WSOC/OC line bisects the graph.
8. Abstract, Figure 4, and elsewhere, “Participation” is stated instead of “Precipitation.”
9. Table 2- Mexico City T0, what is “nan”? If data are not available in the cited publication, measurements are available in other publications.
10. In the text and tables, carbonaceous aerosol concentrations have units of ug C/m³, whereas in the figures they are ug/m³. Please be consistent throughout the manuscript.
11. The authors present a different method for the determination of (OC/EC)primary that minimizes the correlation between EC and OC-secondary, rather than the two more common methods presented (1- using the OC/EC ratio when SOC is minor, or 2- OC/EC from primary emissions inventories). This approach is based on the premise that “OC-secondary and EC were totally from different sources.” However, recent studies have suggested that SOC is a product of the photooxidation of VOC emitted from primary combustion sources, like motor vehicles (Robinson et al., 2007) and biomass burning (Aiken et al., 2008; Grieshop et al., 2009; Lee et al., 2005). Thus, it is expected that there will be some correlation between OC-secondary and EC. In presenting their new approach to estimating (OC/EC)primary, the authors should 1) address the possibility of a relationship between primary combustion sources and OC-secondary, 2) establish precedent for their selected method, and 3) compare their approach to the two more established methods for estimating this quantity.
12. The authors choose to omit OC-non-combustion as a significant source of OC. Typically, OC-non is calculated as the intercept of the OC/EC regression, which in

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Figure 7a is 15.1, suggesting that 15ug/m³ of OC may be from non-combustion primary sources, which corresponds more than half of the observed OC. In light of this result, the authors need to revisit the omission of OC-non from Equation 3.

13. Also, the authors need to comment substantially on the uncertainty in the estimate of (OC/EC)primary and the resulting SOC estimates.
14. Furthermore, how does (OC/EC)primary change under different conditions (i.e. typhoon, vs. local primary influence)?
15. How many data points are included in the EC vs. OC scatter plot (Figure 7a)?
16. Figure 8, WSOC vs SOC, the equation shown here does not correspond to the axes on the graph.
17. Figures 2 and 4 present data that was never discussed in the manuscript, such as potassium, chloride, wind speed, and UVB. If these measurements are not discussed, they should not appear in these figures.
18. Also, a notable omission is CO data in Figure 4, as this measurement is central to the conclusions about OOA and OM sources.

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