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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effects of emission reductions on organic aerosol in the southeastern United States" *by* C. L. Blanchard et al.

Anonymous Referee #1

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Using data collected at the SEARCH network sites from 1999 to 2013, along with ancillary sources, the authors assess the magnitude of various source contributions to observed OC and report and explain observed trends in OC. The topic and result are of great interest and the results surprising. This paper is reporting that much of the OC in the SE US is due to combustion sources, not biogenic SOA, (a topic of much current interest), and that a very large fraction of the combustion component is from biomass burning. Furthermore, they report that the biomass-burning fraction to total OC is increasing and will ultimately dominate OC (again, not biogenic-derived SOA). This is a bold prediction, but is it correct? It all depends on the magnitude of the various sources/components contributing to the OC derived in this study, but all of which are highly uncertain. This uncertainty is not well quantified.

For example, Figure 3 indicates (based on trend lines), that the mean contribution of biomass burning to OC (OCbb/OC) at the CTR site in 2014 is roughly 75%. However, the calculation of OCbb is highly uncertain; an error bar is critical to interpreting this result.

The various data analyses methods employed each have significant limitations and are highly uncertain and a major weakness of the analyses. However, the strength of this paper is that combined, the findings tend to produce a coherent picture. This point could be emphasized more through detailed discussion on how the various analyses are related. Eg, for 2014, as noted above, at CTR OCbb/OC is roughly 75% (figure 3), the factor analysis reported on later in the paper gives OC(combustion)/OC at roughly 60%. Fairly close, but why the difference; which is more believable, do they agree within the uncertainties of each? Support for these two analysis is provided throughout the paper, it is argued that OC(combustion) at CTR is mainly OCbb, since OCbb tracks OC(combustion), which provides more insight on the comparison. In other sections it is noted that OC(combustion) at CTR does not decline, in contrast to other sites, and that EC at CTR is very low, again more insights supporting that most of the OC at CTR is OCbb, similar to the finding of Fig 3. In the current form of the paper, making these connections is not straightforward.

Overall, I recommend publication following the addition of more detailed uncertainty analysis and the inclusion of error bars on all the figures. Furthermore, much better assessment of the various analyses presented here could be made by including comparisons to predictions of other methods made during the SOAS study, such as AMS methods for comparing OA/OC and source apportionment to AMS PMF factors (BBOA etc). Some of these issues are discussed in more detail below.

Specific Comments:

Page 17055, maybe comment on possible reasons why EC ratios between JST and CTR decline by much more in the first and 3rd five year periods compared to OC, if the

OC is related to urban emissions?

Page 17056, lines 17 to 19, might wish to elaborate why would EC, OC and SO4 all be correlated (ie, why EC included) if the OC-SO4 correlation is due to an isoprene SOA formation process linked to SO4?

Section 3.2 OM/OC ratios: - this section should likely include more references, at the least include [Turpin and Lim, 2001].

- define laboratory RH (what is the value); I assume it is the RH at which the filters are weighted. Is TEOM data used here? If so what is laboratory RH in this case?

- what is the uncertainty in the calculation of NM and OM* considering all uncertainties in the calculation. Why is water associated with OC not included? What is the error for not including it? Based on uncertainties, is this really a meaningful analysis? The results seem to suggest so, which is somewhat surprising given the expected level of uncertainty.

- this analyses could be "checked" by comparing to AMS OM/OC data for the SOAS study period.

- it might be noted that a good correlation between OC* and OC in Fig 2 is really not that surprising given that the magnitude of OC* is largely dictated by OC (the y and x axis are closely related).

- possibly, the most important line from the analyses is pg 17058 lines 7 and 8, stating that OC*/OC ratios of 1.6 to 1.9 imply POA is a major source throughout the region. Could one really conclude this if the error associated with the ratio were included? (This is discussed above).

Section 3.3 Biomass-burning tracers

- (Pg 17059, Lines 25 to 29) It is somewhat surprising that (non-soil K) nsK is equated to water-soluble K (K+ ion) by the authors, and both believed to be unique tracers of

biomass burning. Zang et al [Zhang et al., 2010] and others have shown the K+ is not a unique biomass burning tracer. The claim that CTR is unique in this aspect maybe correct, but seems somewhat weak.

- Fig 3 is a remarkable plot, but it is somewhat meaningless without some declaration of uncertainty associated with OCbb. Given all the parameters and assumptions that went into coming up with this value, it may be difficult to assess, but this is a critical issue since the plot suggests that at CTR the average (based on trend lines) ratio of biomass burning OC to total OC is about 75% (using OCbb=1.6 ugC/m3 and OC 2.1 ugC/m3). This does not seem realistic. Furthermore, given the uncertainty, is there really no trend in OCbb in contrast to OC? Maybe some form of statistical test could be performed to assess this more quantitatively, eg paired t-test for differences between OC and OCbb for different periods.

- pf 17061 Lines 19 and 20, give the magnitude of the uncertainty. Ie over or under estimated by how much?

- Why not assess the CTR OCbb with other methods that have been used for computing biomass OC, such as AMS BBOA, for the SOAS study period.

Looking at the scatter plots in the Supp Material it seems to me that this analyses is driven by the large events. This determines the correlations and slopes used in justifying and quantifying the OCbb calculation. Plotting the monthly means in Fig 3 will then also be biased by large periodic events. If this is the case, can one really conclude that generally, biomass burning is a large component of OC and OCbb is approaching OC levels?

- in Fig S9 it may make more sense to compare modern TC to OCbb.

- One might clarify that the method of estimating PM2.5 nsK is based on properties of coarse mode aerosols, ie the ratio of soil K to Si is the same in the coarse and fine modes.

Section 3.4 Principle Component Analyses

This is the main topic of this paper and the most abstruse. Subheadings would make this section easier to follow, eg the authors go through a systematic discussion of each PCA factor; a subheading (each factor name) noting this would be helpful.

Would it help to include in the PCA some of the parameters calculated in the previous sections, eg, OC*/OC ratio, Kbb, OCbb, etc?

Specify more clearly that PCA is carried out at each site, the data was not pooled.

To test if the results reasonable, why not first compare/contrast the PCA results at CTR to the various OA sources identified by other methods during the SOAS study (eg, AMS, specific source tracers, such as IEPOX, etc), then move to a more overall assessment at all sites. This would allow some sense of the differences in the various source apportionment methods and help in the interpretation of the PCA results.

References

Turpin, B. J., and H.-J. Lim (2001), Species contributions to PM2.5 mass concentrations: Revisiting common assumptions for estimating organic mass, Aerosol Sci. and Tech., 25, 602-610.

Zhang, X., A. Hecobian, M. Zheng, N. Frank, and R. J. Weber (2010), Biomass buring impact on PM2.5 over the southeastern U.S.: Integrating chemically speciated FRM filter measurements, MODIS fire counts and PMF analysis, Atm. Chem. Phys., 10, 6839-6853.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 17051, 2015.