

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

Anonymous Referee #2

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This is an ambitious paper that analyzes a unique data set of atmospheric composition across the Southeast U.S. The goal of the Authors is to answer the three science questions that are posed in the Introduction, and the title of the paper “Effects of emission reductions on organic aerosol in the southeastern United States” suggests that the paper will answer all three. The paper, however, leaves me somewhat unconvinced. It reads as a rather elaborate aerosol source attribution exercise, but the uncertainties are large, and in the end I am not sure the Authors have determined what fraction of organic aerosol in the Southeast is controlled by anthropogenic emissions.

Major comments:

A. I think the paper needs to be much more careful about the distinction between an-

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thropogenic and biogenic sources of OC. The paper labels OC that correlates with other combustion products (NO_x, CO, SO₂, sulfate) as combustion OC, thereby leaving the impression that it comes from motor vehicles or biomass burning. Previous work has clearly shown that the formation of secondary organic aerosol in the Southeast takes both anthropogenic and biogenic precursors. For example, it was found during the SOAS campaign that the formation of secondary organic aerosol from isoprene is controlled by sulfate (Xu et al., 2015). Also, nighttime formation of SOA takes nitrate radicals formed from anthropogenic emissions and biogenic VOCs (Rollins et al., 2012). Would the Authors call these sources combustion OC due to the correlation with sulfate and NO_x, respectively? I think the Authors need to acknowledge the interactions between anthropogenic and biogenic sources more clearly, and define up front what nomenclature they are going to use in their source attributions and what exactly the different sources stand for.

B. I think that the Authors missed some opportunities to improve their analysis by not using the SOAS data that were collected at the CTR site in summer 2013. For example, in section 3.2, the Authors use the difference between measured PM_{2.5} and the sum of measured PM_{2.5} species to calculate OM. Such an analysis has large uncertainties. However, for 6 weeks in summer 2013, the measurements to test these assumptions were made at CTR. The second example is the estimate of biomass burning OC using potassium measurements in section 3.3. The Authors admit that the uncertainties in this analysis are large, but do not compare their results against independent estimates of biomass burning OC at CTR during SOAS (Washenfelder et al., 2015). The authors may argue that such analyses would extend the scope of the paper too much, but since the uncertainties in their current analyses are large, I strongly encourage them to make use of the unique data set provided by the SOAS study for comparison with their results and to improve their analysis.

C. The paper could be better in putting the results and findings in perspective. The Introduction contains almost no references, except one very general overview paper and

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one previous paper from the same group. The Authors should include a brief description of previous studies in the literature that have attempted to address the specific science questions posed in the Introduction, e.g. (Carlton et al., 2010; Goldstein et al., 2009; Weber et al., 2007). Also, the SEARCH network is not the only long-term measurement effort of aerosol in the U.S. Data from the IMPROVE networks have been used to derive long-term trends in OC (Attwood et al., 2014; Hand et al., 2013) and there is also the extensive data set from the EPA's chemical speciation network. Source attributions of aerosol in the Southeast have been made using molecular tracers (Kleindienst et al., 2007) and aerosol mass spectrometry measurements (Hu et al., 2015; Jimenez et al., 2009). It would be good to acknowledge the presence of these data and analyses, and compare quantitatively where possible.

D. In Section 3.4, the Authors describe the results of performing principal component analyses on the data. This section is very difficult to understand and appreciate. It is clear that the Authors have done a lot of work to verify their results, but the section gets more and more difficult to read as the text progresses. I would encourage the Authors to shorten this section and convey the outcome of all this work more concisely. The issue is not helped by the way these results are graphically represented. The only graph with results of this analysis that made it into the paper is Figure 4, which shows the trends in the different source contributions over time. To gain confidence in this analysis, the composition of the different factors is equally important. These are presented through Tables 2-11 in the supplemental material, but there is way too much information there for the reader to absorb. Some insightful figures to compare the consistency (or absence thereof) of the different PCA factors across sites, years and (perhaps) different versions of PCA would be very helpful to better appreciate the results from all this hard work.

E. Section 3.5 reads as a summary of some previous papers from this group that compared long-term trends in aerosol species with the trends in emissions. Ultimately, it is not very clear what findings in this section resulted from the present study. This

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section provides the link between source attribution of aerosol and emissions, and is as such important to justify the title of the paper. I encourage the Authors to make it clearer which results are new here, versus a summary of previous work. An additional point is that in all of these arguments, it is assumed that organic carbon responds linearly to reductions in precursor emissions. Studies of ozone chemistry have shown that ozone reductions are anything but linear in precursor emissions, and there is no reason to believe that secondary organic carbon will be different.

Detailed comments:

Table 1: What do the multiplication signs in the EC and OC columns mean?

Page 17056, lines 4-11: There are more recent references on OC/EC ratios from vehicles that would be good to include here. For example, a recent paper on trends in fuel-based emissions estimates is useful as it delineates between gasoline and Diesel vehicles (McDonald et al., 2015).

Figure S2: I wonder what the Authors make of the correlations between sulfate and EC, since they mostly come from different sources: power plants versus biomass burning and motor vehicles sources. The sulfate-EC correlations are almost as strong as the sulfate-OC correlations. I suspect a lot of the correlation is driven by synoptic conditions, i.e. everything decreases during rainy and/or windy periods. That would imply though that the Authors should be more circumspect in using the OC-SO₄ correlations to attribute the OC to combustion sources.

Section 3.2: As mentioned above, using the difference between measured PM_{2.5} and the sum of measured PM_{2.5} species to calculate OM is associated with large uncertainties. The analysis is interesting, but to use the results to derive OM/OC ratios and draw conclusions about sources may be a step too far. An added complication is that the range in OM/OC ratios from biomass burning sources overlaps with those from secondary organic aerosol, so the ratio can be expected to have only limited use for source attribution. If the Authors would like to maintain this section, I think they need

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to add a realistic analysis of all the uncertainties involved and limit their conclusions to the extent allowed by the data.

Section 3.3: The Authors present a nice analysis of aerosol potassium and use the findings to estimate biomass-burning OC from the correlation with non-crustal potassium. The analysis is well done, but the uncertainties in biomass-burning OC are admittedly large. As in the previous comment, I think the Authors should better quantify and propagate the uncertainties in this analysis.

The quality of figures in the supplemental material is much lower than the figures included with the main text. Fonts and font sizes are very inconsistent. Some axis labels and captions are much too small to read. Units are missing in many graphs. Figure S5 has the appearance of a screen shot. The supplemental material is actually very significant as it provides important background information. I am hoping the Authors will take the time to improve the presentation quality.

Page 17061, lines 1-14: The results show a strong seasonal cycle in the concentration of retene at the BHM and JST SEARCH sites. The Authors appear to argue that this may be due to differences in fuel types and dismiss the retene data as an indicator of biomass burning. Retene must have a short chemical lifetime with respect to OH radicals, so the much more efficient chemical removal of retene in the summer could be an alternative explanation for the seasonal cycle.

Page 17065, lines 9-23: The principal component analyses the Authors use, reveal some correlation between biogenic VOCs and combustion-related species. The discussion of these observations is not very convincing. The correlation is almost certainly driven by the seasonality and diurnal variation in emissions, the dynamics of the boundary layer and synoptic conditions, which affect different short-lived species in the same way.

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