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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characteristics, seasonality and sources of carbonaceous and ionic components in the tropical Indian aerosols" *by* C. M. Pavuluri et al.

Anonymous Referee #1

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This paper provides measurements of organic (OC) and elemental carbon (EC) in southern India and provides insights regarding the origin (primary or secondary, biomass, fossil fuel combustion, etc) of this PM10. This is particularly important work because (as they show) organic matter is the most abundant PM10 species, more abundant than sulfate in this region. Additionally, elemental (or black) carbon concentrations are sometimes substantial - a major consideration with respect to radiative forcing. And measurements of this kind in this region are lacking.

A real strength of this paper is that the measurements have been divided up into three time periods where the air sampled has quite similar back trajectories.

However, some substantial improvements are warranted:

I. The results section should be reorganized around the major important findings of the paper using ALL appropriate analyses for each including information about sources along each of the 3 trajectories. The paper has separate sections to present the concentrations, and the mass fractions, and correlations with tracers, etc. Many of these sections discuss what this information tells us about potential source contributions. This makes the paper much longer than necessary and leaves the reader trying to put the big picture together without enough help from the paper.

I see the following major points:

1. Species mass fractions, concentrations and comparison with other measurements in the region. **A major point that is never stated is that OM is the largest contributor to PM10 mass, larger than sulfate.

2. How much is primary vs secondary, water soluble vs insoluble.

3. What do the data tell regarding the source contributions and source regions for Chennai carbon?

4. What can we learn from the ion balance. The authors state that the ion balance suggests the aerosol is somewhat acidic in the winter. What does it mean that the cation to anion ratio in the summer is substantially greater than 1? Does it suggest that ammonia is associated with organic acids?

II. With respect to source contributions. The discussion in the text shows the authors considerable knowledge and talent in interpreting field measurements. They do a very good job of explaining the variety of possible interpretations. In one section the authors use source tracers to aid with this effort. These analyses might benefit from more thought. Correlations between organics and various source tracers are used as evidence that these sources are important contributors to OM. However, it must be acknowledged that meteorology is the main driver of changes in concentrations

and therefore all PM components (whether they come from the same source or not) are likely to be correlated. Components might also be correlated because they come from the same source region but not the same source type. This must be acknowledged!! Perhaps a stronger analysis would be multiple linear regression of OC on several source tracers and possibly also on an indicator of mixing height.

III. Methods. The authors need to specify how much water was used in the measurement of WSOC. WSOC is an operational definition. If more water is used, more OC will dissolve. Accuracy and precision should be provided for all types of analyses. No correction was made for the adsorption of organic gases on the filter. This should be noted.

IV. Estimate of secondary organic aerosol via the EC-tracer method: This method has some important limitations and the potential that these limitations affect the SOC estimates in this paper need to be explored. First, the primary OC/EC ratio can differ substantially from source to source. The authors should consider whether changes in source contributions could introduce substantial bias in their SOC estimates. Particularly there are 3 points on Fig 8b that are much higher than the rest. Are there other explanations besides secondary formation for these points? Estimated SOC is higher at night than day. This could be true, but are the authors confident that this is not because of a local nighttime source with a higher OC/EC ratio? They are probably the largest contributors to the authors' SOC estimates. The authors say that the minimum OC/EC was used to represent primary OC. What was this value? Was it determined separately for the three types of sampling days or was one value used for all days? How different would SOC estimates be if another reasonable approach was used? I feel that the SOC estimates provided are pretty uncertain and the authors need to do more work to convince me that they are good. I note that temperatures are high both in summer and winter, so it is not unreasonable to have secondary formation in both seasons. If secondary OC is a substantial contributor to all samples, primary OC/EC will be overestimated and SOC will be underestimated.

V. WSOC can come from biomass combustion as well as secondary formation. The authors need to be clear about this. Is there a way to apportion WSOC between these two?

VI. The authors conclude that biomass/biofuel combustion is the major source of atmospheric aerosols in this region. I do not doubt that it is a contributor and it may be the largest contributor, but I do not think the authors proved this. They seem to say this because OC is correlated with biomass tracers. However, probably all PM species are correlated due to meteorology. Are the correlations stronger than for other source tracers? If there were good tracers for all sources, multiple linear regression could be helpful.

VII. Details: 1. page 3947 first paragraph - authors argue MSA might be from biomass burning because air mass "not originated from oceanic region in winter" but clearly this air mass was transported over the Bay of Bengal right before reaching the sampling site.

2. page 3948 paragraph starting on line 9 - the conclusions of this paragraph are unsubstantiated. Just because concentrations of OC and EC are lower, does not mean they come from a different type of source. It could be that OC and EC from fossil fuel combustion in northern India are reduced by dilution by the time these trajectories reach Chennai.

3. page 3948 - "clear diurnal" is a little exagerated.

4. page 3951 - "biomass burning is a significant source of EC, OC, SO42-, NH4+, MSA..." I am not sure this paragraph proves this. Perhaps the SO4 in this air mass (trajectory) comes from the same region but instead from coal combustion? Perhaps MSA is added to the air as it is transported over the Sea of Bengal.

5. page 3953 line 25 - WSOC/OC is also high when influenced by biomass burning.

6. In two places in the manuscript the authors state that SO42- was the most abundant

ion. This is true, but it sounds like the authors are saying that it was the most abundant PM10 species, which it was not. It is more important to say that OC was the most abundant species. The authors never say this.

7. Importantly, it looks like OC and EC values in some lines of Table 2 are switched. They do not match the OC/EC ratios in the table. Table 1 should be double checked as well.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 3937, 2011.