Responses to Referee #1 Comments

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.

Response: The authors appreciate the reviewer's comments on this manuscript. Temporal variations of elemental carbon (EC) and organic carbon (OC) (please see Fig. 2a and b, respectively) are comparable in winter but they are different in summer. Interestingly, source tracers (nss- K^+ and levoglucosan) used in this study also showed the similar trends (please see Fig. 1SI given below). These temporal trends suggest that the types of sources that contributed to Chennai aerosols from subcontinental regions (ME/IN and SEA) in winter are same or similar and those from oceanic region (AS/IO) in summer are different. Hence, we decided to discuss the results in terms of two seasonal categories only; winter and summer. However, we discussed the results of mass fractions in terms of three seasonal categories (section 3.3 in the revised manuscript) in order to show the differences in source strength between ME/IN and SEA regions.

Regarding source information in each section, we believe that it is important to discuss the probable reasons following the results in each section in order to make it easier to the reader to understand the trends.



Fig. 1SI. Temporal variations of (a) nss- K^+ and (b) levoglucosan in the tropical Indian aerosol (PM₁₀) samples (n = 49) collected on day- and night-time bases in winter (23 January to 6 February) and summer (22-31 May), 2007 from Chennai, India. Open and solid circles show the day- and night-time concentrations, respectively. Levoglucosan data is from Fu et al. (2010).

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.

Response: As we did not estimate the % contribution of inorganic content (sea salt and mineral dust) to PM_{10} , we did not focus on comparison between organic and inorganic matter. But we gave the average concentration of OC (9.4 µg m⁻³; n = 49) and SO₄²⁻ (6.9 µg m⁻³; n = 49) in the text (please see page 10, line 215 and page 11, line 254 in the revised MS) that shows the higher contribution of OC than SO₄²⁻ to PM₁₀. However, we stated that OC was the most abundant component in PM₁₀ in the revised MS (please see page 2, lines 21-24 and page 10, lines 217-219).

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

Response: We included POC concentrations in the text (please see page 10, lines 224 & 225) and Table 1 and its temporal variation in Figure 2d in the revised MS. We also compared their quantities in the revised MS (please see pages 10 & 11, lines 232-234).

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

Response: The discussion is improved in section 3.1 in the revised MS. The obtained data indicate that biofuel/biomass burnings are the major sources of the Chennai aerosol carbon (please see pages 10 & 11, lines 227-249). The seasonal changes in carbonaceous and ionic components together with air mass trajectories imply that the biomass burning emissions are higher in SEA than other regions (please see page 15, lines 356 & 357).

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?

Response: Higher cation to anion ratios in summer suggests that the cations, including NH_4^+ , might be associated with organic acids. This point is included in the revised MS (please see page 13, lines 301 & 302).

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.

Response: We agree with the reviewer's view that meteorology could influence the changes in concentrations of aerosol components to some extent. However, we believe that correlations between components could mainly be driven by sources rather than meteorology and source region.

Based on the obtained data set and their comparison with the literature we found that biofuel/biomass burnings are the major sources of Chennai aerosols although we do not preclude the influence of other sources: fossil fuel combustion and marine biota. Further the simple linear regression analyses between carbonaceous components and source tracers; nss- K^+ and levoglucosan, confirmed that the major contributions are from biofuel/biomass burnings. We believe that our interpretations are logical and the conclusions are appropriate.

Unfortunately, we do not have mixing height and tracer(s) data for other sources during the study period and hence, it is difficult to assess the % contribution of every source using multiple linear regression analysis.

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.

Response: WSOC was extracted with 15 ml Milli Q water. No correction was made for the adsorption of organic gases on the filter during sampling. Following the reviewer's suggestion, we included these details and analytical errors for all the type of analyses in the revised MS (please see page 7, lines 137, 140, 152 & 153 and 156 & 157).

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.

Response: We agree with the reviewer's view that the estimation of SOC by EC-tracer method involves some degree of uncertainty depending on the choice of primary OC/EC ratio. Generally the most probable primary OC/EC ratio can be estimated for particular location through a linear fit using selected OC and EC observations, if additional tracers of combustion-related primary emissions (e.g. CO) data are available for the study period, otherwise, minimum value among the observed OC/EC ratios in each season can be used. In this study, we have chosen the observed minimum OC/EC ratio in each season (0.7 and 3.35 in winter and summer, respectively) as we do not have any additional data of tracers of combustion-related primary emissions. OC/EC ratios (0.7 and 3.35) used in this study are reasonable as the concentrations of EC and OC vary depending on the type of biofuel and burn rate (Stone et al., 2010). We included this point in the revised MS (please see page 8, lines 179 & 180). However, we could not preclude certain degree of uncertainty associated with the used OC/EC ratios in this study.

We do not have any other explanation other than secondary production for three outliers observed in summer.

Following the reviewer's suggestion, we included the minimum OC/EC ratios used for each season in the revised MS (please see page 8, lines 178 & 179).

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?

Response: It is not possible to apportion the primary and secondary WSOC. We improved the discussion further by comparing WSOC/OC ratios in Chennai to those from other sites in India in the revised MS (please see page 19 & 20, lines 448-458).

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.

Response: No, based on several evidences including coefficient of determination (r^2) obtained by simple linear regression analyses between OC and biomass tracers provided in this study, we proved that biofuel/biomas burnings are the major sources of carbonaceous aerosols in South and Southeast Asia. As we noted earlier, meteorology can influence the concentrations of aerosol components and thus correlation between them for some extent, however, source is the main driver for good correlation.

Unfortunately, we do not have tracer data for other sources and hence, it is not possible to run multiple linear regression analysis.

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.

Response: If marine emission is only the source of MSA, its concentrations should have been higher in summer than winter as the air masses arrived in Chennai originated from oceanic region and also transported over the same region for most of the time in summer (Fig.1). But MSA concentration found to be higher in winter by a factor of 3 than in summer although the air masses originated from continental region, and of course, they passed over the Bay of Bengal for some time (Fig. 1). Thus, it is clear that MSA should has mainly been contributed from other source(s) i.e. biofuel/biomass burning in addition to marine emission.

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.

Response: The lower concentrations of OC and EC in Chennai than in Mumbai, Dhaka and Lahore may not be due to dilution during long-range transport from northern India because black carbon (BC) emission from industrial sources is significant not only in northern India and on the western coast (Mumbai) but also on the eastern coast (between Chennai and Kolkata) and in the southern tip of India (Myol-Bracero et al., 2002). However, we modified the phrase in the revised MS (please see page 14, lines 330-333).

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

Response: We modified this phrase. Please see page 22, line 549 in the revised MS.

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.

Response: Based on the discussion in section 3.1 and 3.2 (probable origin of each species) and seasonal distributions of their fractions as well as the source strength of each region, we concluded in such a way. We can't preclude the influence of coal combustion and marine emissions for certain extent on $SO_4^{2^-}$ and MSA, however, our results show that biofuel/biomass burnings are their major sources in Chennai aerosols. If coal combustion is the major source of $SO_4^{2^-}$ in Chennai aerosols, its concentrations (Table 1) and/or mass fractions should be comparable in all seasons because SO_2 emissions from industrial sources are mainly located along the western coast, in the southern tip, on the eastern coast and northern part of India (Mayol-Bracero et al., 2002), but not (Fig. 5). As already noted in response to comment VII-1, biomass burning is the major source of MSA.

In order to avoid any confusion to the reader, we discussed the seasonal changes in mass fractions in section 3.3 by moving the discussion on diurnal changes (section 3.3 in the discussion paper) to the end as section 3.5 in the revised MS.

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

Response: Yes, it is possible. That is why we considered the primary emissions from biofuel/biomass burnings and secondary production during long-range transport as potential sources based on comparison of WSOC/OC ratios in Chennai with the literature values, including those from other sites in India. Please see page 19 & 20, lines 448-458 in the revised MS.

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.

Response: Following the reviewer's suggestion, we stated that OC was the most abundant component in PM_{10} in the revised MS (please see page 2, lines 21-24 and page 10, lines 217 & 218).

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.

Response: There were typos in Table 2. We corrected them in the revised MS.