

Response to anonymous reviewer # 2:

We would like to thank reviewer for his/her valuable comments. We have tried our best to address the comment point by point.

Note: All the reviewer comments are in normal font and the responses by the authors are in bold and italics.

This study applies PMF analysis to measurements of aerosol particle chemical composition in Kanpur, India during winter 2010. It also describes changes in the microphysical properties of aerosol particles between foggy and clear episodes. This paper is poorly written, poorly organized and confusing. The conclusions are vague and not supported by the data presented in the paper. This paper needs major revisions before it can be published. I will give general comments that I hope will help the authors refocus and better organize the paper. Since the paper needs a major rewrite, it is not worth identifying at this time all the specific instances in the text and figures that need correction.

1) The introduction is wordy and contains numerous inaccuracies (e.g., oligomers are not the same thing as organo-sulfates, accumulation mode particles do not evolve solely from evaporation of cloud droplets). I would suggest deleting the entire introduction, rewrite the rest of the paper and then rewrite the introduction to contain information that is relevant to the information in and conclusions of the paper.

The use of term ‘oligomerization’ was in relation with the acidic nature of the aerosol. The investigators (Gao et al., 2004) have documented abundant formation of oligomer under acidic conditions. The sentence will be rephrased to make the content clearer. The introduction section will be modified by putting the relevant content in view of the relevant suggested changes.

2) This paper needs to be refocused on the PMF results. The data used in this paper has been published previously in Kaul et al. (2011). Therefore, it is not necessary to go into so much detail about the experimental measurements (Section 2). It is also not necessary to present time trends for all the species. Figures 1, 5, 6, 7 could be skipped. The size distributions (Figure 8) were not presented in Kaul et al. (2011) and could be included here, but the time trends (Figure 9) are not useful information.

The repeated content of the experimental section will be referred to Kaul et al., 2011. All the time series plots will be deleted in the revised manuscript. The PMF related results will be moved to main section of the revised manuscript.

3) One of the main points of this paper is the application of PMF to the data, but all the PMF results are buried in the supplementary information. I think these figures should be moved to the main part of the paper.

The suggestion will be incorporated in the revised manuscript.

4) The authors have a basic misunderstanding of what PMF provides. It identifies chemical components that co-vary in time. It does not give sources. If you want to claim that your factors correspond to different sources then you need to provide additional information. For example, does the F1 factor have the same chemical composition as measurements of aerosol particles from a biomass burning source? Does the F3 factor (secondary source) correlate with O₃ or SO₂? Does it correlate with calculated SOA? What do you mean by refractory source (factor F2) and how is it different than mineral dust (F4)? What is your evidence that this chemical composition corresponds with this source? Also, looking at the time trends for the relative contribution of the different factors, there does not appear to be any difference between foggy and clear conditions for any of the factors. This is inconsistent with the conclusion in the abstract that biomass burning aerosols are preferentially scavenged by fog droplets.

A much detailed analysis on PMF has now been performed (Fig 1, 2 and 3 below). In addition to dominant presence of the characteristic species signifying a source in the source profile, the chemical species, which have been used to characterize the sources, are correlated with the corresponding factors to ascertain their emission sources. It was observed that resolving the data into four factors actually splits the refractory source into refractory and dust (as assigned in our submitted supplementary material file) and tracer species of the dust source (mainly Ca²⁺, K⁺) correlated poorly with the corresponding factor. Thus, PMF were resolved into three factors only. The factor F1 was characterized by the presence of WSOC and K⁺ which are the tracer of the biomass burning source; the regression coefficient between F1 and these species were fair (R² ~0.87 and ~0.1 respectively) confirming biomass burning as the source (Fig 1 below). The poor regression coefficient of K⁺ with this factor could be attributed to its preferential scavenging. WSOC and K⁺ both are scavenged which has been documented in our previous study (Kaul et al., 2011). A higher correlation between WSOC and K⁺ further supports their scavenging and the wet removal (Table S7 of the submitted supplementary file). The higher correlation of F1 with WSOC is sufficient to assure its origin from biomass burning as this factor is not contaminated from the secondary source (Factor F3). The characteristic species of the refractory source such as Na⁺, Ca²⁺ and Cl⁻ emitted mostly from brick kilns and power plants (both rely on coal as their energy source) in this region, had fair correlation (R² ~0.93, ~0.30 and ~0.82 respectively) with F2 factor indicating refractory its major source (Fig 2 below). The F3 factor which indicate secondary source, characterized by the secondary species such as NH₄, SO₄ showed fair correlation (R² ~0.91 and 0.50 respectively) with this factor (Fig 3 below).

The correlation between O₃ and SOA is certainly relevant where O₃ is produced upon oxidation of VOCs mostly under dry conditions and subsequently SOA formation takes place. However, in cloud and fogs where aqueous phase produced SOA is prevalent; such correlation could be expected to be poor. For example, in our previous study (Kaul et al., 2011), SOA formation is higher during foggy days whereas O₃ production is reduced during foggy day due to reduced photo-oxidation reaction and reduced incoming solar flux (Kaul et al., 2011). The dissolved oxidizing agents during higher relative humidity conditions seem to play rather important role than the gas-phase chemistry and O₃ production from the gas-phase oxidation of VOCs during dry conditions. Similarly, SO₂ was also scavenged due to wet

conditions and its reduced concentration during foggy days was observed (Kaul et al., 2011). A regression of secondary factor (F3) with O_3 , SO_2 and SOA was performed and poor correlation was observed which could be due to the above mentioned reasons.

Considerable amount of WSOC is coming from the biomass burning (Fig 1 below) and relative contribution of factor F1 gradually increases toward February. The scavenging and wet removal of WSOC is documented by Kaul et al., 2011 due to scavenging and wet removal by the falling fog droplets and droplets impacting onto the leaves of trees and plants. Thus, the reduced relative contribution of F1 during foggy episodes and before 1st week of February could be due to scavenging and wet removal of biomass generated aerosols.

The changes in figures and some of the additional details will be included in the revised manuscript.

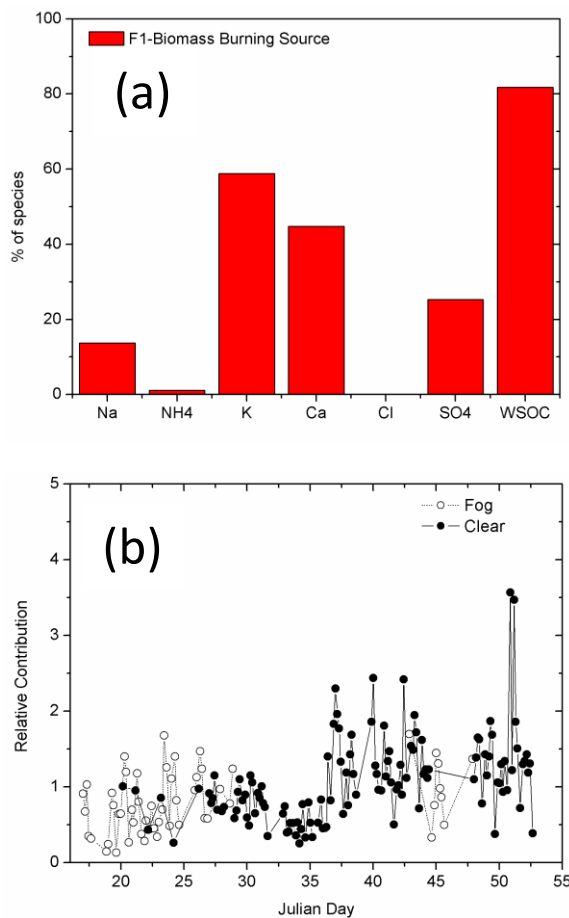


Fig 1- (a) Composition profile of species by factor F1-biomass burning during the study period (b) Time series of relative contribution of F1 during foggy day and nonfoggy (clear) episodes. WSOC is abbreviation of water soluble organic carbon.

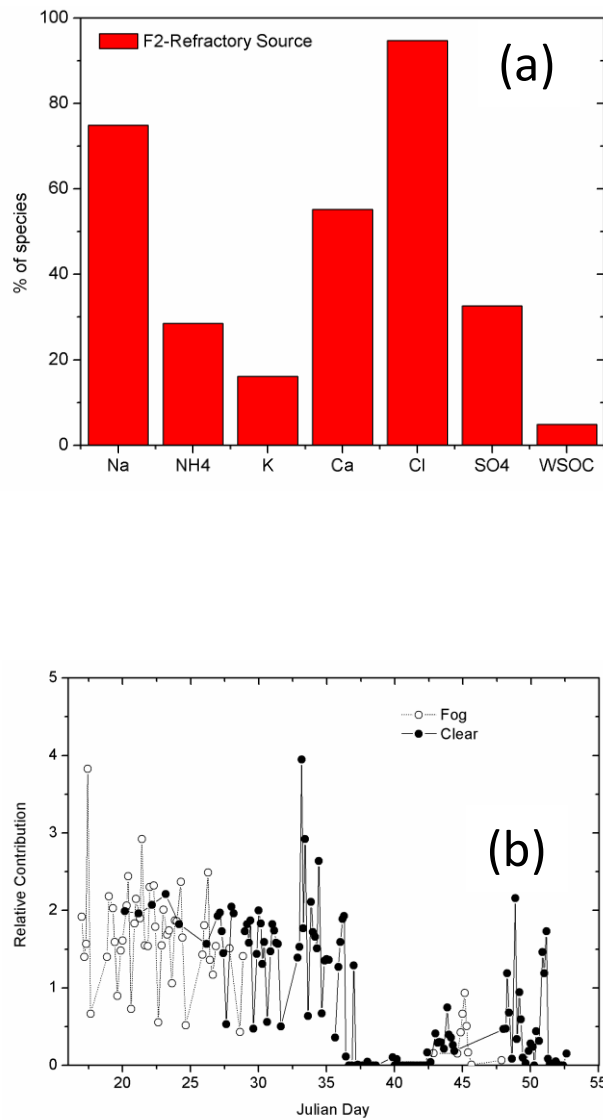


Fig 2- (a) *Composition profile of species by factor F2-refractory source during the study period* (b) *Time series of relative contribution of F2 during foggy day and nonfoggy (clear) episodes. WSOC is abbreviation of water soluble organic carbon.*

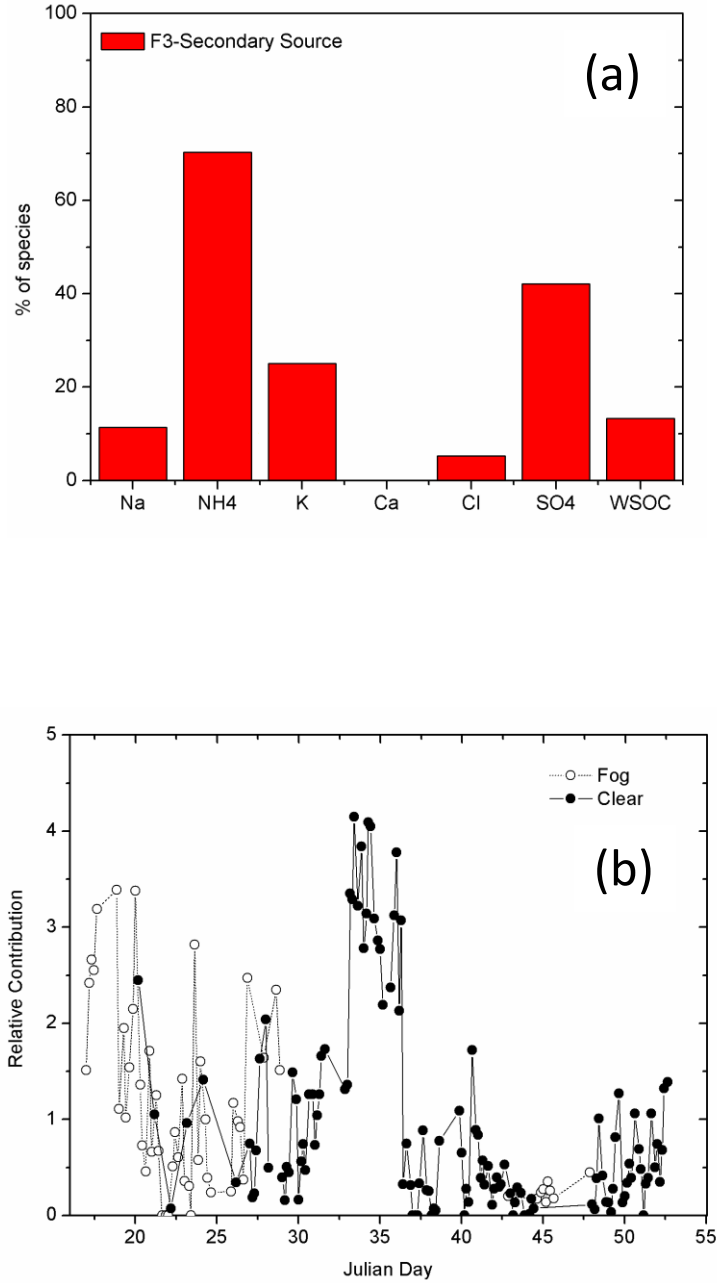


Fig 3- (a) *Composition profile of species by factor F3-Secondary source during the study period* (b) *Time series of relative contribution of F3 during foggy and nonfoggy (clear) episodes. WSOC is abbreviation of water soluble organic carbon.*

5) Figure S9 is difficult to interpret. Use the same x-axis for both panels and use the same log y-axis for both panels. It would be helpful to add a line at $y=1$ to show the change from droplet growth to droplet evaporation.

The subplot of the figure S9 shows the zoomed in part of the figure S9 (Fig 4 below) showing the diameter growth rate for certain diameter range (for diameter > 100 nm). This figure will be modified to make it clearer. A line at $y=1$ will also be inserted in the revised figure.

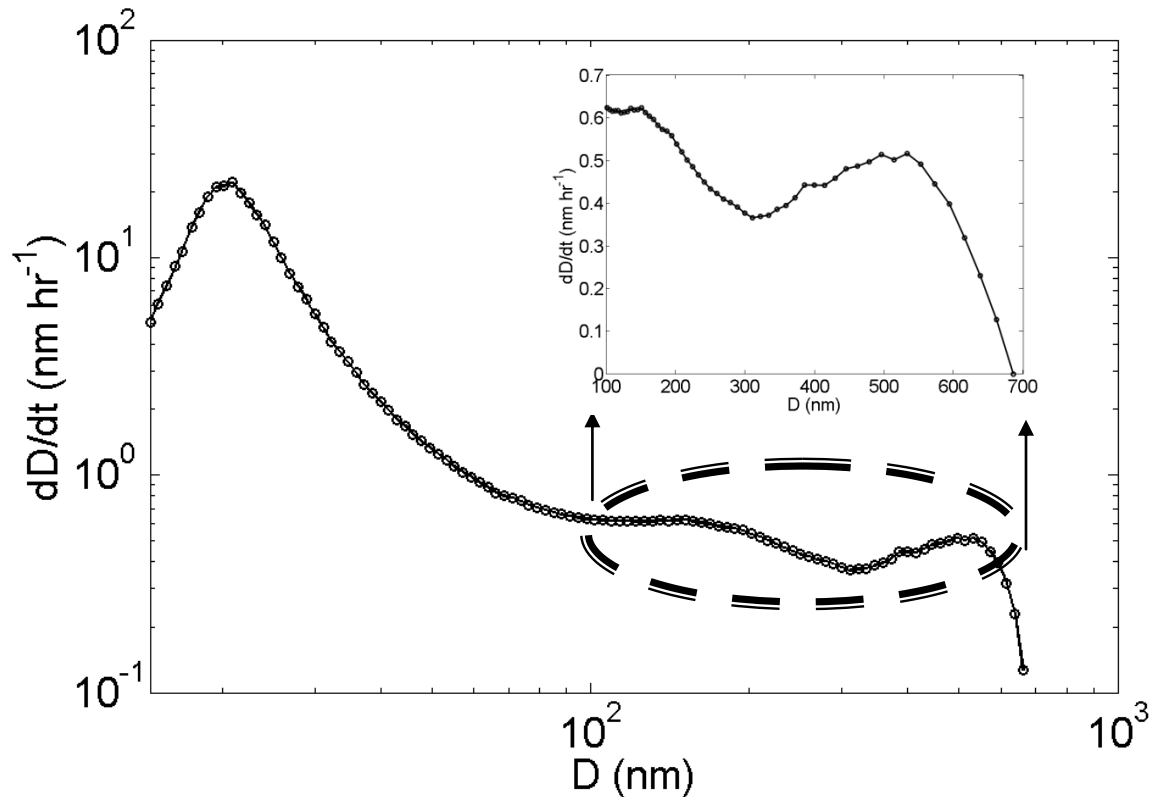


Fig 4- The study average particle diameter growth rate of aerosol during foggy day from overnight (~ 2.6 LST) to 9 LST (a) and from 9 LST to 12.3 LST (b) which are the period of fog evaporation and aqueous phase formation of secondary organic aerosols

6) In Figure 8, the foggy episodes have a higher total particle count and a larger particle diameter. This is not consistent with the statement in the text (Section 3.3) that PM1 mass loading was lower during foggy episodes (although the foggy and clear mass loadings are well within each other's error bars).

To observe difference in the PM₁ mass concentration during foggy and clear episodes, t-test was performed and statistically significant difference (p value ~ 0.018) was observed between

foggy and nonfoggy PM₁ mass concentration. The higher modal diameter (Fig 8-b of submitted manuscript) is attributed to condensation of water over the particles. The evaporative loss of such collected water on the sampled filters during filter storage or during weighing processes could have led to the reduced PM₁ mass concentration. In addition to above, the scavenging and removal of the particles inside the droplets due to falling droplets and droplets impacting onto the leaves of trees and plants could be another reason for the reduced PM₁ mass concentration. The particles inside the droplets are also biomass burning generated as signified by the higher concentration of WSOC and higher regression coefficient between biomass tracers K⁺ and WSOC of the fog water samples (Table S7 of the submitted supplementary file).

7) Once the basic structure of this paper is corrected, it needs to be carefully edited by a native English speaker. There are many incorrect and extremely awkward usages of the English language that make the paper even more confusing than it might otherwise be. Another source of confusion is that the authors do not clearly state when they are presenting chemical composition of aerosol particles and when they are presenting results for collected fog droplets. The source of the data needs to be clear in each paragraph and in each figure caption.

The suggestion regarding the chemistry of aerosol vs. fog water will be included throughout the revised manuscript to avoid confusion. We will try our best to improve the language of the revised manuscript to make the scientific content simple, clear and easy to understand.