Response to anonymous reviewer # 1:

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.

General comments

1) What are your conclusions on the role of aqueous phase processing in modifying aerosol? While you state in the abstract that it changed the mean diameter of aerosol size distributions, throughout the manuscript your findings keep changing and you ascribe increase in aerosol mass to varying boundary layer heights or possibly some aqueous phase processes.

Aqueous processes caused growth in size of the aerosol upon fog evaporation which was due to secondary organic aerosol formation. Fog also removed those aerosols which were highly hygroscopic such as biomass generated aerosols. In the abstract, it was mentioned that the modal diameter (not mean) of the number size distribution increases and that is attributed to secondary aerosol formation mainly upon fog evaporation and also during fog formation periods (Fig 8-b of the submitted manuscript). The primary aerosols such as those produced from vehicular and industrial emission will accumulate during reduced boundary layer conditions, mainly during overnight, early morning and evening.

2) The use of the term 'interstitial particles' is misleading. Interstitial particles are usually those that are not activated into droplets. I have the impression that the authors imply that 'interstitial particles' are the particles that acted as condensation nuclei and are immersed in droplets.

By this word, we meant particle immersed in the droplet. The word will be replaced with "immersed in the droplets" and will be corrected throughout in the revised manuscript.

3) There seems to be a conceptual misunderstanding of SOA formation in the aqueous phase. The authors discuss at several places that inorganics (ammonium, sulfate) are required to form SOA mass in the aqueous phase. The processes that are referred to here are all processes that occur in the aqueous phase of very concentrated, deliquesced aerosol particles. Chemical processes in the aqueous phase of much more dilute fog droplets do not require any inorganics. There is a huge body of literature that shows that e.g. oxalic acid or related acids are formed by such processes (e.g., (Crahan et al., 2004; Sorooshian et al., 2006; Ervens et al., 2011)). Both oxalate and sulfate are formed in droplets and thus appear in the same mode (droplet mode) but there is no chemical interaction required.

Most of the secondary organic aerosol formation is due to partioning of organic compounds such as oxalic and related acids into organic phase as supported by (Crahan et al., 2004; Sorooshian et al., 2006; Ervens et al., 2011). The interaction of inorganic species with organic compounds and subsequent formation of organosulfate and organonitrate in aerosols and clouds is also a part of the secondary organic aerosol formation which has been shown by various investigators (references at lines 3 through 5 of page 14488 of the submitted manuscript) and Tolocko et al., 2012. The inorganic species such as ammonium and sulfate could help in more oligomer formation in the aerosol (Gao et al., 2004).

Although, Lim et al., 2010 have not provided any threshold value of concentration of organic mass per liter of fog water for occurrence of such interaction inside the fog droplets. The study average organic carbon mass content of the fog water in large, medium and small fog droplets during our sampling period was $40 \pm 25.6 \text{ mgC/l}$, $98.2 \pm 101 \text{ mgC/l}$ and $423.8 \pm 479.8 \text{ mgC/l}$, respectively which indeed indicates considerably large mass of organic carbon inside the fog droplets (Kaul et al., 2011). These details are in Kaul et al., 2011 and at lines 25 through 27 of page 14499 and at lines 1 though 5 of page 14500 of the submitted manuscript. These concentrations values will be referred to Kaul et al., 2011 in the revised manuscript in support of the fog water concentrated with organic and inorganic species (Fig 4 of the submitted manuscript).

The acidic nature of aerosol (references at lines 16 through 24 of page 14487 of the submitted manuscript) has produced more organic compounds and possible causes includes enhanced partioning and subsequent formation of hetero-molecular compounds or enhanced oligomer formation (Gao et al., 2004) which are though mostly limited to chamber studies and are not very well understood. Finally, chamber studies are conducted to mimic the actual atmospheric phenomenon under controlled conditions. Certainly, oxalate and sulfate are formed inside the droplets without any chemical interaction but there are numerous studies (discussed at lines 1 through 5 of page 14501 of the submitted manuscript) which has documented chemical interactions. Thus, additional studies are required to ascertain the actual mechanism and in this regard, our study will be useful to the scientific community. If indeed there is no interaction, the values of organic mass provides to the scientific community a threshold below which their formation may be negligibly small; although, the better correlation of organic carbon of the fog droplets with the inorganic species does indicate such interaction (Table S7 of the submitted manuscript). A recent study on ambient aerosol (Tolocko et al., 2012) has also estimated the contribution of organosulfate and has found that it constitutes ~ 5-10% of the organic mass. It also cannot be claimed with certainty about their independent formation either. Although, the evaporating fog can act as aerosol water because species present inside the droplets becomes concentrated due to droplet evaporation. Thus, a positive correlation between organic carbon and inorganic species (possible sulfate and nitrate) of the aerosol could be expected but such correlation was poor.

Some of the above discussion will be included in the revised manuscript.

4) Given the fact that the authors conclude that SOA formation in fog might have been rather negligible and that individual compounds have not been identified anyway, the review-like text in the introduction of detailed processes can likely be condensed. In fact it seems that the authors have heavily used the review article by Ervens et al., 2011, ACP, for this text. Thus, it can be significantly shortened with the appropriate reference.

The above conclusion is misinterpreted. We have shown in our previous article (Kaul et al., 2011) that the enhanced formation of secondary organic aerosol takes place due to aqueous

phase chemistry during fog episodes and upon fog evaporation. In this study, we have tried to understand the role of inorganic species in the formation of secondary organic aerosols, mainly the formation of organosulfate and organonitrate and enhanced oligomer formation in acidic conditions. We report that organosulfate and organnitrate which are also part of the SOA could have likely formed inside the fog droplets but their formation was negligibly small in the aerosols; their absence in the aerosol could be due to their overnight removal by the fog droplets. These details were includes at lines 1 through 22 of page 14501; lines 18 through 23 of page 14506 of the submitted manuscript.

Some of the additional details will be included in the revised manuscript.

5) Unfortunately you do not show any concentrations of VOCs for the duration of the measurements. Could differences in aerosol composition (partially) ascribed to different emissions?

Emission sources remained the same during both clear and foggy day and throughout the study period as implied by the presence of the tracer species, although their intensity may have changed. If emission sources had changed then we should have observed the presence or absence of the tracer species of that source but such trend was not seen. The atmospheric conditions and aqueous chemistry is the major cause of the different aerosol composition.

6) The source apportionment seems to have been associated with large uncertainties (e.g., K+). In addition, the four identified factors are mostly characterized by species rather than by specific sources. Some discussion is needed in order to link these species to sources.

A much detailed analysis on PMF has now been performed (Fig 1, 2 and 3 provide 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, have been correlated with the corresponding factors to ascertain their emission sources. It was observed that resolving into four factors splits the refractory source into refractory and dust (as assigned in our submitted supplementary) and tracer species of the dust source (mainly Ca^{2+} , K^+) did not correlate with that 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 (R^2 ~0.87 and ~0.1 respectively) indicating its biomass combustion origin (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^+ of the fogwater further supports their scavenging and the wet removal (Table S7 of the submitted supplementary). The higher correlation of F1 with WSOC is sufficient to assure its biomass source as this factor is not contaminated from the secondary source (Factor F3). The characteristic species of the refractory source such as Na^+ , Ca^{2+} and Cl emitted mostly from the brick kilns and power plants (both sources rely on coal as energy source) in this region, had fair correlation (\mathbb{R}^2 ~ 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 NH_4^+ , SO_4^{2-} showed fair correlation ($\mathbb{R}^2 \sim 0.91$ and 0.50 respectively) with this factor (Fig 3 below).





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 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 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.

Specific comments

p. 14485, l. 6/7: reword this sentence.

The sentence will be rephrased as "Aerosol acts as cloud condensation nuclei for fog formation. The stable condition, low wind speed and low mixing height accompanying the fog causes accumulation of the primary particles in the fog layer" in the revised manuscript

p. 14485, l. 17: how does sulfate get attached to BC?

Ulevicius et al., 1994 and other references there in have documented that heterogeneous SO_2 oxidation on black carbon particles during high humidity and inside the fog droplet and subsequent mixing of sulfate with black carbon was found to be important in the atmosphere; during winter time, 90 % of the sulfate was found to be mixed with black carbon. The sentence will be rephrased and some of these additional details will be included in the revised manuscript.

p. 14485, l. 25: that is very vague: how can scavenging be identified by measuring the listed constituents?

This sentence will be rephrased to make it clearer.

p. 14487, l. 20: There are several studies that show that SOA in the aqueous phase might be composed of photochemically produced compounds without any heteroatoms, e.g., (De Haan et al., 2009; Lim et al., 2010)

It is answered in reply to comment number 3 and 4.

p. 14488, l. 3-5: Most of these references do not refer to ambient studies as implied by the text.

The references listed are both from ambient and chamber experiments. Chamber experiments are carried out to better understand the actual process in the atmosphere. Documenting the findings from both chambers as well ambient studies will help in understanding the actual mechanism in the atmosphere rather than relying on only chamber experiments. In that perspective, ambient measurements acquire greater significance.

p. 14488, l. 6: how does SOA formation in the aqueous phase change number concentration? (here and at other places throughout the manuscript)

We have analyzed the size distribution data of every day in detail and no new particle formation event was found. The concentration is mainly due to primary particles. The sentence will be corrected in the revised manuscript.

p. 14488, l. 7: do you mean surface and volume distributions?

Yes, these are surface and volume size distributions. They will be corrected in the revised manuscript

p. 14489, l. 1: There are numerous studies that have explored the absorptive properties of SOA, e.g., (Shapiro et al., 2009; Trainic et al., 2011; Nguyen et al., 2012)

This sentence is irrelevant to this work and will be deleted in the revised manuscript.

p. 14492, l. 17: O3 is barely soluble. Has the study by Herrmann et al (or others) indeed shown that the aqueous phase can be an efficient sink for O3?

The lesser concentration of O_3 during foggy day is due to reduced photo-oxidation reaction (Kaul et al., 2011). Small amount can also diffuse inside the droplets which could be the other reason of its reduced concentration during foggy episodes and these studies (Hermann et al) have documented the same. The meteorological section, as suggested will be deleted in the revised manuscript and will be referred to Kaul et al., 2011.

p. 14493, l. 17: Can you give a rough estimate (<1%, <10%, 50% : : :?) of how much uncertainty was introduced by these additional factors?

Uncertainty was estimated as contribution of the additional factor divided by the sum of all the contributing factors. It was found that uncertainty for different species were less than 20 %. This information will be included the revised manuscript.

p. 14493, l. 28: Does the fact that all species were categorized as 'weak' mean that they concentrations were roughly on the same order of magnitude. Some explanation might be useful here for readers who are not familiar with PMF.

The concentrations were not the same order of magnitude as can be seen from the Fig 1 through Fig 9 of the submitted manuscript. The procedure for assigning a species as 'weak' or 'strong' is based on signal to noise (S/N) ratio as documented in the references (line 29, page 14493 of the submitted manuscript) and same procedure was adopted. Guidelines related to the signal to noise (S/N) ratio for assigning the species such as strong or weak depends upon the relative S/N ratio of the other species. In the strongest variable, the error is minimal where as in the weakest variable the data could entirely be noise. It was recommended (Paatero and Hopke, 2003) that the species having S/N ratio greater than 2 could be assigned as strong, those having S/N ratio less 2 and greater than 0.2 could be assigned as week and those having S/N ratio less than 0.2 could be assigned as bad. Following their recommendations, all the species were categorized as weak in our study.

The additional details will be included in the revised manuscript.

p. 14494, l. 8ff: If the aerosol is dominated by biomass burning – shouldn't PMF simply show one factor? Why were these period excluded? How certain is your source apportionment considering these large uncertainties in K+?

These concentrations, if plotted as time series with the other data points, will be observed as outlier. These five data points which had very high concentration were considered as outlier. Such rare events are also not captured by the PMF as the source profile indicates the average source contribution to each sample. Thus, a few aerosols which are predominantly affected by single source will not be reflected in the average source profile and source contribution to each sample.

The species which are removed from the system where source apportionments is carried out would not be apportioned correctly and thus its actual concentration could not be tracked. The additional details will be included in the revised manuscript.

p. 14494, l. 21: what does the refractory factor tell you about possible sources?

The possible sources are brick kilns and power plants, vehicular and industrial emissions and this information will be added in the same line. The sources over this region have been well identified and are included in references at lines 25 through 25 of page 14489 of the submitted manuscript.

p. 14495, l. 6: can you speculate on what this large identified fraction might have included?

Metals and their oxides have not been identified and quantified which can possibly contribute to this unidentified fraction.

p. 14495, l. 10-13: This sentence combines two completely different facts ('ionic species affect visibility and 'organic mass formation in the aqueous phase')

This sentence will be rephrased to make it clearer. Organic aerosol formation also changes the chemical, optical and physical properties of the aerosol which is documented by Trainic et al., 2011.

p. 14495, l. 26: NH4+ is not oxidized – it simply dissolves. NH3 is usually taken up by droplets in order to neutralize the present excess of anions.

We also meant the same i.e. NH_3 is taken up by the droplets and NH_4^+ is subsequently formed by the oxidation of NH_3 . This sentence will be deleted in the revised manuscript as the same information is provided in the introduction section.

p. 14496, l. 1: do you mean that sulfate and ammonium can be directly emitted from specific sources?

The sulfate can come from $CaSO_4$ salt which could be emitted from the refractory (possibly brick kilns and coal power plant). Possible source of sulfate from biomass could be due to the

dust deposited on the leaves which could be re-suspended upon fire and captured at the sampling site. The origin of NH_4Cl to biomass could due to its formation by reaction of ammonia and chloride both emitted from the biomass. The presence of various salts in the fresh and aged smoke is documented elsewhere (Liu et al., 2000; Li et al., 2003). Some of these details will be included in the revised manuscript.

p. 14496, l. 7: what does the 'higher water solubility' refer to? (higher than what?) Did Pratt et al. identify the same factor as you did in your study? Do you have any measurements of hygroscopicity (e.g., growth factors or hygroscopicity based on AMS–derived composition) that would support your conclusions on the higher solubility of biomass burning aerosol as compared to the other factors? How much does the hygroscopicity differ between the factors? Is it significant and sufficient to cause any effects on water condensation and fog properties (e.g. droplet number)?

Biomass burning generated fresh and aged aerosol are hygroscopic compared to the aerosol generated from other sources such as refractory. We do not have access to AMS which limits us to provide any detail on hygroscopicity of the aerosol emitted from the various sources. Although, numerous articles have documented the hygroscopic nature of the biomass burning generated aerosols. We are indirectly referring the hygroscopicity and wet removal of biomass generated aerosols by looking at the trend of water soluble organic carbon (WSOC) and K^+ concentrations (Kaul et al., 2011) and relationship between them (this study).

p. 14496, l. 14: There are many studies that show this drop size dependence and discuss sizedependent sulfate formation rates. Some of the references should be added here, e.g., (Collett et al., 1994; Rao and Collett, 1998)

These suggestions will be incorporated in the revised manuscript.

p. 14497, l. 22: What do you mean by secondary production of Ca2+, Na+ and NH4+? These are primary species that are not formed in the atmosphere.

We also mean the same i.e. Ca^{2+} and Na^{+} are primary species and NH_{4}^{+} is secondary. The time series plots and discussion pertaining to them, as suggested by other reviewer 2, will be removed in the revised manuscript.

p. 14497, l. 22ff: Figure 5 shows that these species rather increase during fog. I suggest highlighting and discussing this fact rather than implying that these species were removed by the fog and thus decrease upon dissipation (or is this what you mean?)

Species are removed overnight by larger fog droplets. It can be seen from Fig 4 of submitted manuscript that smaller droplet are more enriched with these species than the larger ones which has comparatively lesser removal rate. Thus, these smaller droplets which still remain suspended in the atmosphere after sunrise, when evaporate, will cause increase in concentration of the concerned species in the aerosol. The concentrations of ammonium, sulfate and nitrate after their overnight removal have increased upon fog evaporation as shown in Table S8 of submitted supplementary. The higher study average concentration of

these species during foggy day may be attributed to accounting such increase upon fog evaporation in the average concentration. Both processes i.e. wet removal mainly during overnight and their formation through aqueous phase process could be the cause of such trend. The additional discussion will be included in the revised manuscript.

p. 14498, l. 27ff: are you implying that it is actually not the fog but different emissions that causes differences in aerosol composition?

Emission sources remained the same during both clear and foggy day and throughout the study period as implied by the presence of the tracer species, although their intensity may have changed. If emission sources had changed then we should have observed the presence or absence of the tracer species of that source but such trend was not seen. The atmospheric conditions and aqueous chemistry is the major cause of the different aerosol composition.

p. 14499, l. 6ff: I don't understand this section. Sulfate and nitrate are both nearly completely scavenged, i.e. the H2SO4 concentration is negligible in the gas phase whereas there might be small concentrations of HNO3 indeed present. The hygroscopicity of NH4NO3 and (NH4)2SO4 is comparable.

The NO_3 and HNO_3 (g) is effectively scavenged and removed whereas SO_4^{2-} and H_2SO_4 (g) are poorly scavenged because of lower water solubility of SO2 (g). This finding is documented in Aikawa et al., 2007 and this article is referred at line 3 of page 14486 of the submitted manuscript. The sentence will be rephrased in the revised manuscript to make the sentence clearer.

p. 14499, l. 24: This header should be reworded according to my general comment 3).

This header will be slightly reworded to reflect the relevant content. More details are included in answer to comment number 3.

p. 14499, l. 3 (and Table S6a): How does an single value give information about processing? It would be more meaningful to compare values at the beginning and at the end of a fog event.

These are not single values but average concentrations of several fog water samples. That is why standard deviation is also included in this table. Fog usually starts from 6:00 pm and persist until mostly 01:30 pm next day. The fog had also persisted for continuous 48 hours on few days. Fogwater was not collected at different time of a fog event and thus such comparison could not be carried out.

p. 14500, l. 17: An increase of SOA (OC) in smaller droplets might point to formation processes that occur in a concentrated aqueous phase as often referred to as 'aerosol water' (Lim et al., 2010). Do the aqueous phase concentrations (organic mass/water volume) support such speculation?

Although, Lim et al., 2010 have not provided any threshold value of concentration of organic mass per liter of water. The study average organic mass content of the fogwater in large,

medium and small fog droplets during our sampling period was 40 ± 25.6 mgC/l, 98.2 ± 101 mgC/l and 423.8 ± 479.8 mgC/l which indeed indicates considerably large mass of organic inside the fog droplets (Kaul et al., 2011). These details were included in Kaul et al., 2011 and at lines 25 through 27 of page 14499 and at lines 1 though 5 of page 14500 of the submitted manuscript. These concentrations values will be referred to Kaul et al., 2011 in the revised manuscript in support of the aerosol water.

p. 14500/14501: The mass accommodation coefficient is not dependent on drop size (unless there is some organic coating that is more enriched on smaller droplet which would lead to a decrease of this coefficient). However, the phase transfer rate is inversely proportional to drop size

The inorganic carbon and related content will be removed in the revised manuscript as discussion pertaining to them is irrelevant to this study. The organic carbon content of the fog droplets will be referred to Kaul et al., 2011.

p. 14501, l. 14: Your previous study (Kaul et al., 2011) reported on clear SOA formation in fogs in the same region and time. What is different in your current study that you conclude that SOA formation in fogs was negligible?

In our previous study we have documented enhanced SOA formation due to aqueous phase process in which numerous organic compounds such as carboxylic acids partition into the organic phase. A part of SOA formation is through heteroatom molecular compounds such as formation of organosulfate and organonitrate which also contributes to the SOA formation. Our findings reports their possible contribution inside the fog droplets whereas negligible contribution in the aerosols, possibly due to their overnight removal. More details regarding these are included in section 3.5 of the submitted manuscript.

p. 14502, l. 16: Are you implying that new particles form during fog events? This cannot happen in fog droplets since each droplet already includes one (or more upon scavenging) particle and additional mass is imply added. Is there any evidence from lab and/or field studies that enhanced RH – such as during fog events - facilitates new particle formation?

We have analyzed the size distribution data of every day in detail and no new particle formation event was found. The concentration is mainly due to primary particles. The sentence will be corrected in the revised manuscript.

p. 14502, l. 16: Separate clearly here which effects are due to changing boundary layer and which ones can be ascribed to particle growth? Some guidance could be possibly given by {Eck, 2012 #2985}.

We have analyzed the size distribution data of every day in detail and no new particle formation event was found. The higher concentration during foggy episode is mainly due to accumulation of primary particles during reduced boundary layer conditions. The sentence will be corrected in the revised manuscript. p. 14503, l. 3: what is meant by 'leave aqueous oxidized organic compounds behind that form new particles'? New particle formation is usually referred to as the process of forming small clusters of gas molecules (e.g. H2SO4 or possibly organics). These particles have sizes of a few nanometers. – Is this indeed the process you refer to?

We have analyzed the size distribution data of every day in detail and no new particle formation event was found. The concentration is mainly due to primary particles. The sentence will be corrected in the revised manuscript.

p. 14503, l. 23: Could the fact that you see poor correlation of particles < 40 nm with photooxidation be explained by the fact that particles grow to larger sizes and thus are those that indeed show a better correlation?

We have analyzed the size distribution data of every day in detail and such growth was not observed.

Figure captions: There seem to be random numbers in all figure captions (e.g. 4, 5 in Figure 1).

They will be corrected in the revised manuscript

Figure 1: It might be helpful to add the total mass to the pie charts.

In addition to the percentage of the species, the mass concentration of the species (in μgm^{-3}) is also annotated in the same figure. The revised figure is provided below (Fig 4 below) and will be included in the revised manuscript.



Fig 4 - (a) Aerosol chemical composition during foggy episode (b) Aerosol chemical composition during nonfoggy (clear) episode. POA, SOA and EC stands for primary organic aerosols, secondary organic aerosols and elemental carbon, respectively. The values in the closed bracket are concentrations in μgm^{-3}

Figure S2-5: What does the % refer to? I tried to add up the contributions from the single compounds form the four factors or adding up the contributions of all species within each factor and none of them seems to make sense (> 100%).

These are the study average percentage of the species present in the aerosol. We have rechecked these values and they make up to 100%.

Table S7: What was the contribution of the three drop size classes to the total drop populations? Could there be some statistical issue that biases the correlations?

Since, number size distribution of the fog droplets were not measured, contribution of these droplet size classes to the total droplet population is unknown and thus statistical issue could not be commented on.

Figure S9 and S10: I assume that unit on the axes should be nm. Could you show an additional figure that shows the evolution of the size distribution over a foggy period (i.e. two curves: initial and processed size distribution)?

The correction regarding unit will be done in the revised manuscript. Additional figures showing average size distribution at 19.25 hour local standard time (LST) (less processed) and 22.30 hour LST (more processed) during foggy episode is shown as below (Fig 5 below).



Fig 5- Fog processed aerosol number size distribution at 19.25 and 22.30 hours local standard time (LST). D stands for aerosol mobility diameter.

Technical comments p. 14490, l. 11: 'surface' instead of 'diameter' or 'diameter of 2.11 cm'?

It is surface area (~ 2.11 cm²); Most of the content of experimental section will be deleted and will be referred to Kaul et al., 2011 as suggested by reviewer 2.

p. 14491, l. 29: Fig. 1?

All the time series plots refer to fig 1 through fig 9. All the time series plots will be deleted in the revised manuscript as suggested by reviewer 2.

p. 14503: semi-VOC should be 'semivolatile VOCs'

The suggestion will be incorporated in the revised manuscript.

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