

We are thankful to the two referees for thoughtful comments which help improve the manuscript substantially. Following the reviewers' suggestions, we have revised the manuscript accordingly. Listed below are our point-by-point responses in blue to each comment that is repeated in italic.

### **Response to Referee #1**

*This paper reports an observational case study of pollution in Beijing around the APEC period, chiefly with an HR-AMS. The techniques used are fairly well-established, but the results presented are extremely interesting because this presented a unique opportunity to study the different sources of megacity pollution in isolation. Several interesting observations are made regarding the different sources of PMF-resolved organics and the changes in the oxidation state of the organics and the sizes of the particles. It is generally well written and very relevant to ACP, so I recommend publication after the comments below have been considered.*

We thank the reviewer's positive comments.

*This is not the only paper covering this case study; the paper Chen et al. ([www.atmos-chem-phys-discuss.net/15/22889/2015/](http://www.atmos-chem-phys-discuss.net/15/22889/2015/)) also covers this from the perspective of the measurements made on the Beijing Meteorological Tower. While I have read both papers and I am satisfied that there is not too much overlap between the two papers, I find it strange that this paper makes no reference to the Chen et al. paper, especially as the two papers share the same corresponding author. It would be useful to discuss the relevance of the findings presented here in the context of the observations of the other paper.*

Thanks the reviewer's comments. Chen et al. (2015) was now cited more in the revised manuscript for a better comparison with the results obtained at the ground site. For example, in section 3.1, we added "However, the measurements at 260 m at the same location showed significant decreases of 40–80% for all aerosol species during APEC, whereas the bulk aerosol composition was relatively similar before and during APEC as a result of synergetic controls of aerosol precursors over a regional scale(Chen et al., 2015). These results indicated the different sources of aerosol particles between the ground site and 260 m."

*General: The use of the term 'oxidative properties' seems a little odd. This is normally used to refer to the properties of oxidizing agents or their precursors (e.g. NO<sub>x</sub>, O<sub>3</sub>), but here it is the oxygen content of the organic aerosol that is under investigation. I think it would be more correct to refer to the 'oxidation properties' throughout the manuscript (as it is done in a few instances).*

A good point. “oxidative properties” was changed to “oxidation properties” throughout the manuscript.

*Page 23411, line 5: The statement that ‘the oxidative properties of aerosol particles remain largely unknown’ is a little odd because there have been a large number of papers focusing on this exact topic in the last seven years using the techniques used here. While it continues to be a subject of much interest, I don’t think the statement as it is written really stands.*

Thank the reviewer’s comment. This sentence was now revised as “the oxidation properties of aerosol particles remain less understood”.

*Page 23415: The terms ‘A-A’ and ‘I-A’ should be defined.*

Thanks the reviewer’s comments. The AMS organic O/C tends to be biased low because of the influence of unimolecular ion decomposition reactions, in which a fragment with an electronegative atom such as oxygen has a larger tendency to become a neutral, rather than a cation. The study by Aiken et al. (2008) showed that the bias can be accounted for by dividing an average calibration factor (0.75) which was obtained from laboratory standards. However, a recent study found that the O/C values from the method of Aiken et al. (2008) are systematically biased low, particularly with larger biases for alcohols and simple diacids. Therefore, a new method on the basis of O/C from Aiken et al. (2008), fraction of  $\text{CO}_2^+$  and  $\text{CHO}^+$  was proposed for the calculation of O/C of organic aerosol (Canagaratna et al., 2015). To differentiate these two methods, the method of Aiken et al. (2008) was defined as “Aiken-Ambient” (A-A), and the one proposed by Canagaratna et al. (2015) was defined as “Improved-Ambient”. Following the reviewer’s suggestion, we defined A-A and I-A in the revised manuscript.

*Page 23418: The discussion regarding aerosol acidity is almost certainly not correct. If the aerosol was acidic, it would be unable to support nitrate, which would partition completely to the gas phase as nitric acid. Reports of acidic aerosols in the atmosphere measured using AMS (e.g. in marine locations) always feature very little or no nitrate and show much worse correlations than is presented in figure 2. It is far more likely that in this instance, the aerosol is pH neutral and the sub-unity  $\text{NH}_4$  meas/pred value is because sulphate RIE used here is inaccurate. This parameter can and does vary from the default value of 1.2 and should have been calibrated along with the RIE of ammonium.*

Thanks the reviewer’s comments. The default RIE (1.2) was used in this study for sulfate quantification. Unfortunately, we didn’t calibrate the AMS with pure  $(\text{NH}_4)_2\text{SO}_4$  particles. As the reviewer mentioned, the RIE of sulfate might be a factor affecting the calculation of the predicted  $\text{NH}_4^+$ . In addition, a considerable fraction of

biomass burning was observed in this study which can emit chloride in the form of KCl. As a result, we may also overestimate the predicted  $\text{NH}_4^+$  by counting all chloride as  $\text{NH}_4\text{Cl}$ , and hence overestimate aerosol particle acidity. The cations such as  $\text{K}^+$  and  $\text{Na}^+$ , and the organic acids that the AMS cannot quantify will introduce further uncertainties in evaluating aerosol particle acidity. For these reasons, we deleted the discussions on aerosol particle acidity in the revised manuscript. We also deleted the section of aerosol particle acidity in Chen et al. (2015) which likely has the same issue.

*Page 23420: Could the variation in the HOA/BC ratio also be caused by changes in the relative contributions from the local combustion sources such as biofuel, coal and traffic?*

Thanks the reviewer's comment. Coal combustion was not a major source of organic aerosol during this study for two reasons: 1) the heating season in Beijing started from November 15 which was behind this study; 2) PMF analysis of high resolution mass spectra of organic aerosol did not resolve a factor associated with coal combustion emissions, while it can be resolved during the heating season (Sun et al., 2013). The reduction of BBOA (16%) during APEC was similar to that of HOA (19%), and the ratio of HOA/BBOA was 0.81 during APEC, which is also similar to that (0.84) before APEC. Therefore, we infer that the variations of HOA/BC ratios could not likely due to the changes of relative contributions of local combustion sources.

*Haze event: Please use a different numbering convention than 'S1', 'S2', etc. for the stages because these are also used for the supplement figures and I found this confusing.*

Thanks the reviewer's comment. The four stages were now defined as "E1, E2, E3, and E4" in the revised manuscript.

*Figure 13: The vertically-resolved wind speed and direction data needs to be properly introduced. Is this the same data as was used in figure 14a of Chen et al.?*

Yes, it is the same data as that in Chen et al. (2015). Following the reviewer's suggestion, we added a description of the wind profile measurements in the revised manuscript. "The wind profiles including WS and WD between 100 m to 5000 m were measured by a Doppler wind lidar (Windcube 200, Leosphere, Orsay, France) at the same location."

*Figure 14: The caption needs to be specific about what event is being shown in this Figure*

We revised the figure caption as:

Fig. 14. (a) Evolution of size distributions of sulfate, nitrate, and organics during the severe haze episode between October 22 and 25 (Fig. 12). (b) Average size distributions of sulfate, nitrate, and organics during the four stages of E1-E4.

*Supplement: Are the ion tracers referred to in S1 and S2 derived from AMS data? If so, these are internal, not external tracers.*

Good point. They were from the AMS measurements. We deleted “external” in the captions of Figs. S1 and S2.

## Response to Referee #2

*The manuscript reports detailed chemical characterization of PM<sub>1</sub>, mostly NR-PM<sub>1</sub> during an important event (APEC) in Beijing. Strict atmospheric legislation emissions were imposed during APEC, which gave the authors the opportunity to investigate aerosol properties and sources before and during APEC, including severe haze episodes. The authors demonstrate experience on the techniques and provided very detailed information. Although not many new insights are presented, it seems to me that the results have implications on atmospheric chemistry and for this reason I recommend it to be accepted for publication in this journal after the considerations below are taken into account.*

Thanks the reviewer's comments.

*General comments:*

*Language was often poorly written, I recommend careful text revision, especially with the use of 'plural' form.*

Thanks the reviewer's comments. We had a careful proofread in the revised manuscript.

*In the introduction the authors claim that 'understanding of size distributions and elemental compositions of OA in Beijing remains poor'. Nowhere in this planet there have been so many studies with HR-AMS including HR-PMF, size distributions and elemental composition of OA till the present moment (I counted 8 studies!). No doubt that they were all important. However, I do not believe such sentences can be used as motivation statements.*

Thank the reviewer's comments. HR-AMS has been widely used in field campaigns for characterization the size distributions and elemental composition of organic aerosols. Here we refer to the HR-AMS studies in Beijing. To our knowledge, the number of reported HR-AMS ambient studies in Beijing is approximately 9 (Huang et al., 2010a;Huang et al., 2010b;Liu et al., 2011;Zhang et al., 2014;Elser et al., 2015;Sun et al., 2015;Zhang et al., 2015a;Zhang et al., 2015b;Zhang et al., 2015c). Although several of these studies discussed the size distributions and elemental composition, most of them focused on the average size distributions for the entire study period and/or presented the time series and diurnal cycles of elemental ratios. Few of them discussed the evolution of size distributions during the severe haze episode, and also the evolution of oxidation degree of OA as a function of relative humidity was rarely investigated. Therefore, our understanding of the size

distributions and elemental composition of OA is still limited. Following the reviewer's comments, we revised this sentence as: "our understanding of the evolution of size distributions and elemental composition of OA in Beijing is still limited."

*The authors say that emissions control during the Olympic Games 2008 were implemented to a lesser degree. Please provide more detail on that.*

The emission controls during the Olympic Games included temporary closures of factories and restrictions on traffic by alternating the odd and even plate numbers. The control measures were mainly implemented in Beijing, while similar but less extensive traffic and industry restrictions were imposed in Beijing surrounding areas within a radius of about 150 km (Cermak and Knutti, 2009). In the revised manuscript, we added a more detailed description of the emission control during the Olympic Games and cited the reference of Cermak and Knutti (2009) for further details.

*Limits of detection for the HR-AMS were estimated and presented, however, it surprises me that some compounds presented much lower values than the ones presented by DeCarlo et al. (2006), especially ammonium. Do the authors have an explanation for that?*

There are several reasons for such differences: 1) the detection limits we reported are 5-min averages while those reported in DeCarlo et al. (2006) are 1-min values; 2) the detection limit of HR-AMS is instrument dependent. For example, the HR-AMS in DeCarlo et al. (2006) was the early version, while the performance of HR-AMS has been significantly improved during the last 10 years; 3) the ambient environment, e.g., relative humidity, might also be a factor affecting the detection limits, particularly ammonium, because the fragments of ammonium (mainly  $m/z$  16, and 17) have interferences from water fragments ( $O^+$  and  $OH^+$ ). We removed the statement "which are close to the values reported in previous HR-AMS studies (DeCarlo et al., 2006)." in the revised manuscript for clarification.

*I suggest the authors include the input organics matrix dimensions, so the readers will know the number of variables and samples used in the PMF analysis. Also, explain if PMF was run for all the period (before APEC, during APEC) together or separately.*

Good point. We clarified this information in the revised manuscript as:

“The PMF2.exe algorithm (v4.2) in robust mode (Paatero and Tapper, 1994) was applied to the HRMS matrix (4158×306) of OA for the entire study period to resolve distinct OA factors representing specific sources and processes.”

*In section 3.1 the authors say that nitrate contribution was larger than in previous studies. Is that an indication that legislation imposed during APEC was more effective for this compound? Why?*

The higher nitrate contribution than those in previous studies indicate that nitrate played a more important role in PM pollution during the study period. One of the major reasons is the continuous reduction of SO<sub>2</sub> in recent years while the NO<sub>x</sub> level remained at a high level and even increased. Another part of the reason is because of the season (autumn) for this campaign, when the temperature is not high enough to evaporate NH<sub>4</sub>NO<sub>3</sub> substantially while the photochemical production is still strong (Zhang et al., 2012b). Based on our analysis, the emission controls over a regional scale during APEC showed similar impacts on nitrate and sulfate, and it is difficult to tell that the legislation imposed during APEC was more effective for nitrate reduction.

*Concerning the acidity, do the authors have additional information concerning cations, such as K<sup>+</sup>, Na<sup>+</sup>, Rb<sup>+</sup>? For example, Takegawa et al. (2009) measured those ions with tan AMS in Beijing. In an environment where mass loadings are so elevated, one could expect them to be relevant for the neutralization.*

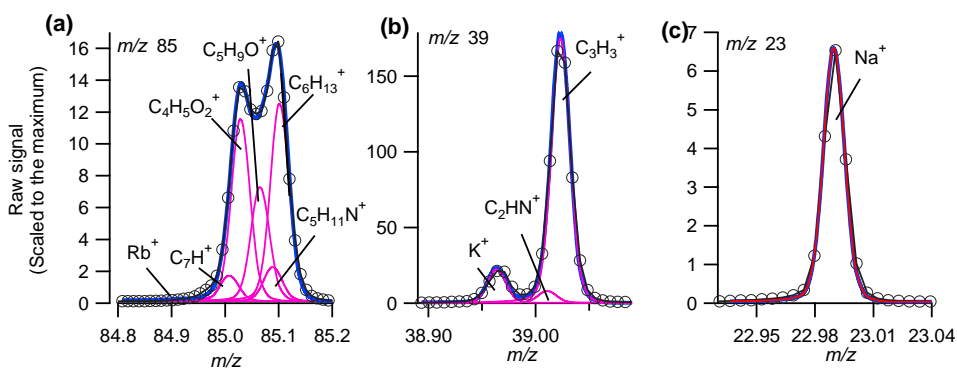


Figure R1. The raw spectra of (a)  $m/z$  85 (b)  $m/z$  39 and (c)  $m/z$  23

The high resolution mass spectra showed clear signals of K<sup>+</sup> and Na<sup>+</sup> (Figure R1). However, quantification of K<sup>+</sup> and Na<sup>+</sup> is challenging due to the surface ionization

issue and also the unknown RIE of  $K^+$  ( $RIE_K$ ). The  $RIE_K$  can vary a lot depending on the tuning of the spectrometer and the temperature of the vaporizer. For example, Slowik et al. (2010) reported a  $RIE_K = 10$  based on the calibration of pure  $KNO_3$  particles using a ToF-AMS, which is much higher than the  $RIE_K = 2.9$  obtained from the comparisons of K/S from fireworks and AMS measurements (Drewnick et al., 2006). In addition, the stability of surface ionization (SI) and electron impact (EI) also affects  $RIE_K$ . For these reasons, we didn't quantify  $K^+$  and  $Na^+$  with the HR-AMS measurements.

The  $K^+$  and  $Na^+$  will affect our evaluation of aerosol particle acidity in this study. For example, a considerable fraction of biomass burning was observed in this study which emits chloride mainly in the form of  $KCl$ . We may overestimate the particle acidity by counting all chloride as  $NH_4Cl$ . As the reviewer #1 mentioned, the RIE of sulfate and the organic acids may introduce further uncertainties in calculation of aerosol particle acidity. Therefore, we deleted the discussions on aerosol particle acidity in the revised manuscript.

*Diurnal cycles of SIA components were very similar, increase in the afternoon. To me that suggests mixing layer development. Regional pollutants stay imprisoned above the mixing layer during the night and during the day when the mixing layer evolves they are released down to the surface level.*

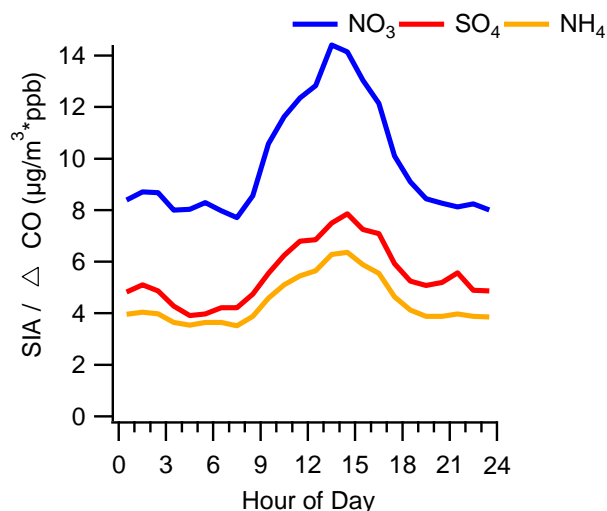


Figure R2. Diurnal cycles of SIA/ $\Delta CO$  ( $\Delta CO$  is the CO minus background CO).



As stated, planetary boundary layer (PBL) played an important role in changing the diurnal cycles of SIA components. Because of the stronger convective turbulence and higher temperature, the boundary layer height during daytime is generally much higher than that at night. The rising boundary layer would dilute the concentrations of pollutants leading to lower concentrations during daytime. Figure R2 shows the diurnal cycles of SIA/ $\Delta$ CO which can remove the dilution effect of PBL to a certain degree. It is clear that secondary inorganic species showed pronounced daytime peaks indicative of daytime photochemical production. Therefore, the increased concentration of SNA in the afternoon was primarily caused by photochemical processing. It is possible that the pollutants in the residual layer at night can be mixed downwards to the ground site during daytime. However, we cannot draw such a conclusion based on the current data analysis in this study.

*Concerning the COAs factors, the authors associate COA2 to charbroiling because that was the banned cooking technique during APEC. Could the authors provide further evidence of charbroiling cooking factor? For example, this factor presented  $CHN^+$  and  $CHNO^+$  fragments, especially for larger  $m/z$ s ( $>m/z80$ ). Any insights on what could those fragments represent? How could they be related to charbroiling and not to typical cooking?*

Thank the reviewer's comments. Charbroiling was banned in certain areas, e.g., Huairou district in Beijing during APEC. However the charbroiling activities were still significant in the streets near our sampling site. We currently don't have a good explanation for the high nitrogen-containing ions in cooking aerosols, which should be explored in the future by sampling directly the source aerosol of charbroiling.

Previous GC/MS analysis showed that unsaturated fatty acids were abundant organic aerosol species from Chinese cooking emissions (He et al., 2004; Zhao et al., 2006). The mass spectra of typical cooking organic aerosol are characterized by strong signal at  $m/z$  41 and 55 (He et al., 2010), which is similar to the spectral profile of COA1. Whereas the spectrum of meat charbroiling showed high ion signals at  $m/z$  43 and 55 (Mohr et al., 2009), which was similar to that of COA2 in our study. Based on these comparisons, we infer that COA1 was related to the typical cooking aerosol and COA2 was mainly associated with charbroiling.

*It surprises me that all POA factors (but COA1) contain significant fractions of  $m/z$  60 and 73. Did the authors try ME2 to better separate the factors? They look rather mixed.*

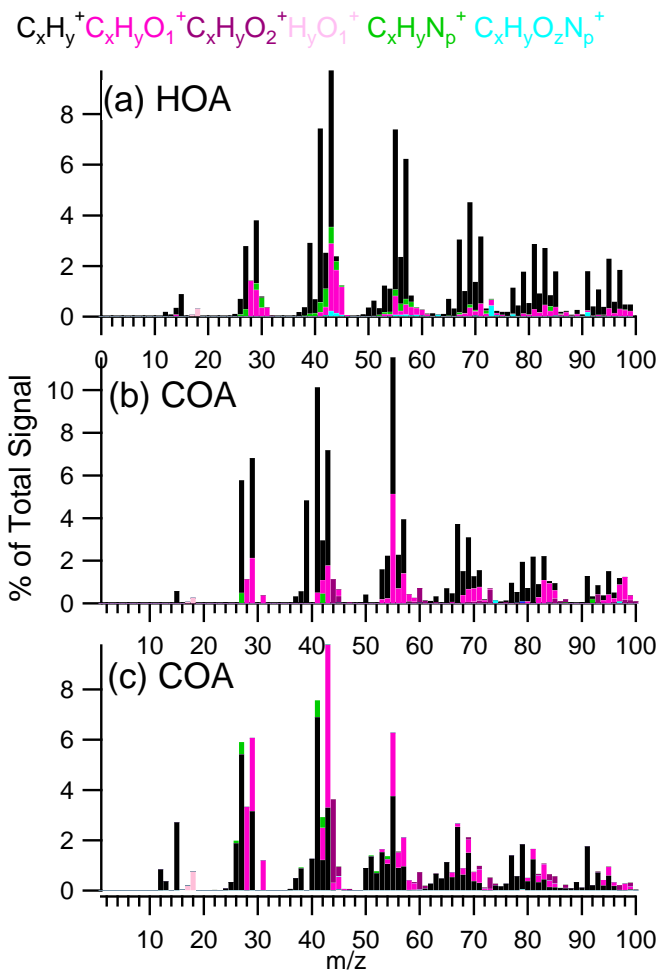


Figure R3. High-resolution mass spectra of OA resolved from previous studies, including (a) HOA (Aiken et al., 2009), (b) COA (Crippa et al., 2013) and (c) COA (Mohr et al., 2012).

Thank the reviewer's comments. We didn't try the advanced ME-2 analysis yet. The high  $m/z$  60 in COA is consistent with that resolved in previous studies, e.g., Paris, 2009 (Crippa et al., 2013), Barcelona 2009 (Mohr et al., 2012), and also those in source profiles of Chinese cooking emissions (He et al., 2010).

The high  $m/z$  60 in HOA is not typical. However, the spectral patterns and the comparisons of time series with external tracers (CO and BC) indicate that the HOA factor was well separated. One explanation is that HOA was mixed with a small amount of coal combustion organic aerosol (CCOA) that cannot be resolved by PMF. The mass spectrum of CCOA is similar to that of HOA, yet shows visible  $m/z$  60 peak

(Elser et al., 2015). However, due to their similar spectra and the small contribution of CCOA before the heating season, PMF analysis cannot resolve the CCOA factor.

*Why was the terminology SV-OOA and LV-OOA utilized instead of OOAI and OOAI (Huang et al., 2010)? Both OOAs are very oxidized ( $m/z$  44>43) and both present similar agreement with nitrate and/or SIA. It is not clear to me that the factor labeled as SV-OOA is more volatile. Concerning diurnal cycle of SV-OOA, how can the mountain valley breeze influence only this factor and not the others? Please explain.*

The terms of “OOAI” and “OOAI” are widely used in early AMS, mostly Quadrupole-AMS studies which refer to highly oxidized and freshly oxidized oxygenated organic aerosol, respectively. Recent HR-AMS studies together with the volatility measurements (Huffman et al., 2009b; Huffman et al., 2009a; Jimenez et al., 2009) suggest that OOAI is characterized by high oxygen-to-carbon (O/C) ratio with low volatility while OOAI with lower O/C is found to be more volatile than OOAI. In addition, many studies found that OOAI often correlated with sulfate and OOAI often correlated with semi-volatile nitrate. Since then, the two types of OOA, i.e., OOAI and OOAI are commonly defined as low-volatility oxygenated organic aerosol (LV-OOA) and semi-volatile OOA (SV-OOA), respectively. In this study, the O/C of LV-OOA (=0.99) is much higher than that of SV-OOA (=0.47). To be consistent with previous studies, we named the two types of OOA as LV-OOA and SV-OOA, respectively. Although the two OOA both showed high  $m/z$  44/43, we noticed a considerable fraction of  $C_xH_y^+$  ions in SV-OOA, further indicating that it is less oxidized than LV-OOA.

The mountain-valley breeze influenced all OA factors as indicated by the diurnal cycles in Figure 6. All OA factors started to decrease from midnight when the mountain-valley breeze occurred.

*SIA and OOA showed similar large accumulation modes peaking at 650nm. Usually, the lens transmission efficiency drops abruptly at  $D_{va}>600$  nm. Please explain how could that be? Was the transmission efficiency measured for this instrument?*

Thank the reviewer’s comments. We didn’t measure the lens transmission efficiency for our HR-AMS. One of the explanations is that there was a considerable amount of SIA and OOA in the larger particles, e.g.,  $D_{va}>600$  nm before APEC. In fact, a recent HR-AMS study using a  $PM_{2.5}$  lens in Beijing (Elser et al., 2015) showed that the size distribution of sulfate during the pollution episode peaked at ~1000 nm while nitrate

showed the highest concentration between 600 – 1000 nm. Our study showed maximum sizes at ~650 nm for SIA which might indicate that the increased concentrations between 600 – 650 nm overcame the decreases caused by lens transmission efficiency. To be accurate, we revised “peaking at ~650 nm” as “peaking at ~650 nm or even larger”.

*In the Krevelen diagram, slopes before and during APEC are very similar (-0.52 and -0.58) to infer that different aging processes took place.*

We agree with the reviewer that the slopes before and during APEC were similar. Therefore, we used “slightly” to describe the differences.

#### *Minor Comments*

*P23414, L13: Write W-mode.*

#### Changed

*P23416, L17: NO<sub>x</sub> or NO<sub>y</sub>? In section 2.1.1 NO<sub>y</sub> was mentioned. Please choose one.*

It is NO<sub>x</sub>. NO and NO<sub>y</sub> were measured by the gas analyzer (model 42i ) from Thermo Scientific. The NO<sub>2</sub> was measured by a cavity attenuated phase shift NO<sub>2</sub> monitor (Aerodyne Research Inc.), which was described in section 2.1.1. The NO<sub>x</sub> was then calculated as NO<sub>x</sub>=NO+NO<sub>2</sub>.

*P23416, L19-22: Sentences need clarification. Were the two SV-OOAs combined? Please, write clarify.*

We reworded this sentence as:

“The five-factor solution yielded a mixed SOA factor and the seven-factor solution split the SV-OOA into two components, which cannot be reasonably explained due to limited external tracers.”

*P23417, L11: 3rd, not ‘3’.*

We changed it to “November 3”.

*P23417, L18: Write ‘composed of’ down.*

Added.

*P23417, L20: Did you mean organic mass fraction?*

Yes, we revised it as “The contribution of organics showed a large increase, accounting for more than half of PM<sub>1</sub>, whereas that of SIA was decreased from 51.2% to 35.4%.”

*P23417, L27: Please report the NO<sub>3</sub>/SO<sub>4</sub> value for Zhang, et al. (2014), so the reader can compare it.*

Added.

*P23419, L7: Spell out BA and DA.*

Spelled out.

*P23419, L23: Please cite few chloride combustion sources.*

Added.

“Chloride, mainly from combustion sources, e.g., biomass burning and coal combustion (Levin et al., 2010;Zhang et al., 2012a), showed similar diurnal cycles before and during APEC”.

*P23421, L4: The authors probably meant ‘increase’, not ‘decrease’. Please correct.*

Corrected.

*P23421, L17-18: Already described in section 2.1.1, no need to mention it again here.*

Removed.

*P23421, L21: The word correlates is not appropriate, replace it by ‘in agreement’.*

Revised as “This result is consistent with the fact that vehicles use was limited only between 3:00 and 24:00 during APEC.”

*P23427, L-14-15: This sentence is not clear.*

It was reworded as “coinciding with a time without traffic control, and likely having more traffic emissions during APEC.”

*P23432, L26: Please include the meteorological effects after emission control.*

Added

*Fig. 5 (f) right panel, there’s a typo, ‘SNA’.*

We changed “SNA” to “SIA”.

Fig. 4 was cited after Fig 5 in the manuscript.

Changed

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