We thank the reviewers for their insightful and useful comments. Our specific response can be found below and are in italics and blue colored font. Changes made to the manuscript are in quotation marks.

Responses to Anonymous Referee #1

This paper describes measurements of aerosol composition made in Fresno, California in wintertime during one of the NASA DISCOVER-AQ campaigns. Time series, diurnal cycles, inorganic/organic composition and sources, organic elemental ratios, size distributions, and source directions of submicron aerosol are presented. PMF was used to investigate source and processing for the organic aerosol. Effects of weekend/weekday, boundary/residual layer mixing and pollution events are discussed. Several meteorological and gas-phase measurements are used to interpret the aerosol measurements. Comparisons are made to a similar measurement dataset/publication which showed some differences such as an additional BBOA factor (with fairly different mass spectra) and OOA factor and shifts in OA factor contributions, and different residual layer effects which is mainly attributed to meteorological differences coupled to possible changes in wood heating burning.

The manuscript is generally well written and describes a clear, straightforward, and thorough analysis of the measurements on a general level. However, like Referee 2, I also found that despite the clearly large effort that went into collecting and analyzing the measurements, there is little new in terms of methods or scientific insight nor are the measurements put in context compared to other such measurements at other locations. This seems to be particularly the case in light of the authors' similar publication of similar measurements and analysis at the same location during a different winter. Many observations are pointed out and then either one or a few possible explanations are briefly put forward usually with little in-depth analysis to investigate and better elucidate the underlying explanations. In short, it reads more like a report of measurements than a scientific study, especially given that new methods are not applied nor is the sample location/season new.

That said, like Referee 2, I would also err on the side of publication since the measurements and data analyses are thorough and high quality, and despite a lack of in-depth scientific analysis, there doesn't appear to be issues with over-stated conclusions that all-too-often find their way into the literature. Moreover, I suspect that documentation of these results will be useful in the context of ongoing DISCOVER-AQ analyses and future investigations and efforts to understand and mitigate the air quality issues of the SJV.

We thank the reviewer for his/her comments, although we disagree with some of the comments on the value of this work. The San Joaquin Valley (SJV) of California is one of the most polluted regions in the US despite many years of regulatory control efforts. There has been much uncertainty surrounding of the sources, both local and regional, and processes of aerosols in the SJV and numerical models perform very poorly at predicting aerosol concentration and properties in this region. Thus there is an urgent need for detailed measurements at high time resolution and the measurements performed in Fresno in 2013 was aimed at addressing this need.

Due to the nature of measurement campaigns, during which intensive, real-time measurements often last only for a relatively short period of time (e.g., a couple of weeks up to a couple of months), the results may not always be representative or applicable to other time periods or years. Comparing measurement results between different years is highly valuable for understanding the influence of emission sources, atmospheric processing, and meteorological conditions on aerosol. For this study, the fact that similar measurements were performed in a nearby location in winter 2010 provides a great opportunity to investigate the influence of meteorology on the aerosol chemistry. As we discussed in the manuscript, the meteorological conditions were significantly different between these two winters – the 2010 winter, due to the influences of the El Nino, was wet and mild with frequent occurrence of fog and rain whereas the 2013 winter was generally dry, sunny, and cold. The meteorological condition in winter 2013 was also more stagnant with low wind speed on average. As a result, daytime photochemical processing in the SJV was significantly more intense during DISCOVER-AQ whereas aqueous phase processing was a more important mechanism for secondary aerosol production during winter 2010.

We published two papers on the 2010 AMS dataset, which was one of the first highly-time resolved aerosol chemistry datasets acquired in the wintertime in the SJV. The Ge et al. (2012a) elucidated the sources and processes of organic aerosols (OA) and highlighted the important, and leading, roles played by anthropogenic primary OA emissions, primarily from vehicular traffic, cooking, and residential heating, in contributing and influencing aerosol pollution in Fresno in the wintertime. The Ge et al. (2012b) paper provided a thorough discussion on the influence of aqueous phase processing on aerosol composition and size distributions. In comparison, in this paper the focus is on meteorological influences on aerosols in terms of their sources, including changes in anthropogenic behavior in response to changes in meteorological conditions, and processes. The present work revealed new insights, particularly regarding the nocturnal residual layer, and we presented new analyses such as analysis and examination of the weekday/weekend differences, polar plots of source regions, and comparisons with the 2010 data set. These results are new and insightful, enabling us to identify the strong influence of meteorology and sources on the aerosols in the SJV as well as the important role of the nocturnal residual layer plays in influencing secondary aerosol concentrations.

Given the focus of this paper is to investigate the aerosols at Fresno measured with high-time resolution and compare the data between two years from this same location, putting these results in the context of other measurements at other locations would not have as much relevance as they may have in other studies, especially regarding the aims of this work. Including discussions of this nature would also likely result in additional lengthening of the manuscript. However, results from other studies at different locations both in the US and Europe, are discussed in this paper, both to aid and support the interpretation of the results from this project such as the elemental ratios of each of the OA factors identified from positive matrix factorization analysis as well as the differences between the two biomass burning OA factors.

More detailed responses are given to the reviewer's specific comments below.

Detailed Comments:

P35059, L11: "This is an indication that nighttime chemistry might also be higher in 2013.": It is not clear what this means. Higher above ground? Faster? What types of chemistry? Please clarify.

In 2013, the concentrations of secondary aerosol species increased more rapidly in the morning than they did in 2010, suggesting that the formation of these species during the night was more intense in 2013 and thus played a more important role in influencing aerosol concentrations at the ground in 2013 than 2010. The reasons include higher precursor gas concentrations in 2013 and stronger influence of a residual layer where nighttime chemical reactions promote the formation of nitrate, sulfate, and OOA. The nocturnal residual layer appeared to be much weaker in 2010 compared to 2013. To clarify, page 35059, line 11 has been amended in the revised manuscript to read: "This is an indication that nighttime chemical reactions may have played a more important role in 2013."

P35060, L23-24: "representing an important PM component" is very vague. Be more specific.

This sentence has been amended in the revised manuscript to read: "Previous studies have shown that the composition of ambient aerosols in Fresno, one of the most populated cities in the SJV, is complex, with organic species representing an important component of PM, often contributing up to two-thirds of the total mass (Chu et al., 2004; Chow et al., 2006; Turkiewicz et al., 2006; Ge et al., 2012a)."

P35061, L2-3:" In addition, the typical cold and wet weather in the winter promotes gas-to-particle partitioning of semi-volatile species." Is this correct? Indeed cooler temperatures would promote higher partitioning of SVOC to the organic phase, but why would wetter temperatures promote gas-to-particle partitioning of SVOC? You mean of soluble compounds?

Gas-to-particle partitioning of semi-volatile species depends strongly on temperature. Temperature is typically anti-correlated with RH thus a combination of colder temperatures and high RH promotes the partitioning of gas-phase species to the particle phase. In addition, wet weather promotes the gas-to-particle partitioning of soluble compounds. However, in the manuscript, the term 'wet weather' was in reference to higher humidity (RH) rather than greater precipitation. For clarification this sentence has been amended in the revised manuscript to read: "In addition, the typical cold and high humidity weather in the winter promotes gas-to-particle partitioning of semi-volatile species."

P35062, Sect 2.1: A reference to a white paper, overview or NASA website would be useful here. Also it would seem appropriate to introduce (mention) the aircraft measurements here.

Following the reviewer's suggestion, we now provide a link to the white paper as well as a link to the main website. In addition, a link to the data that is available for public download is included. The aircraft measurements are also introduced in this section. This section has been amended in the revised manuscript to read: "DISCOVER-AQ is a coordinated effort mission combining surface and aircraft based measurements to characterize and understand how aerosols and trace gases evolve throughout the day and across urban areas. The overarching aim of the DISCOVER-AQ project (http://discover-aq.larc.nasa.gov/) is to relate concurrent observations of column abundances to surface concentrations of key gaseous pollutants and aerosols to improve the interpretation of satellite observations and diagnoses of near-surface air quality (http://discover-aq.larc.nasa.gov/pdf/DISCOVER-AQ_science.pdf). One of the objectives is to characterize the differences in diurnal variation of surface and column observations for key trace gases and aerosols. To achieve this goal, gas and particle-phase measurements were made throughout the day from two aircraft and a network of US ground sites that *experience diverse meteorological and surface conditions. The factors that contribute to local air quality problems (e.g., emissions, transport, and chemistry) also vary between the sites. Of the two aircraft, the NASA P-3B made daytime measurements close to the ground, at constant altitudes of ~2500 m or ~400 m, or flew in tight spirals to measure vertical profiles throughout the SJV. The vertical profile measurements were made over seven select ground locations, including the Fresno supersite where detailed ground measurements were made to allow for quantitative connections to be made between the surface aerosol concentrations and properties and those observed aloft (e.g. Pusede et al., 2016). Data from the DISCOVER-AQ project is available to the public at: http://www-air.larc.nasa.gov/missions/discover-aq/discover-aq.html."*

P35063, L8: Add reference for SP2.

The reference Schwarz et al. (2006) has now been added for the SP2 in the revised manuscript.

P35064, L18: change "low" to "lower". Saying that a high-resolution ToF has low resolution seems contradictory. (also change high to higher).

As suggested by the reviewer, we have changed the relevant words in this sentence in the revised manuscript.

P35064, L20-21: State frequency of calibrations during the campaign.

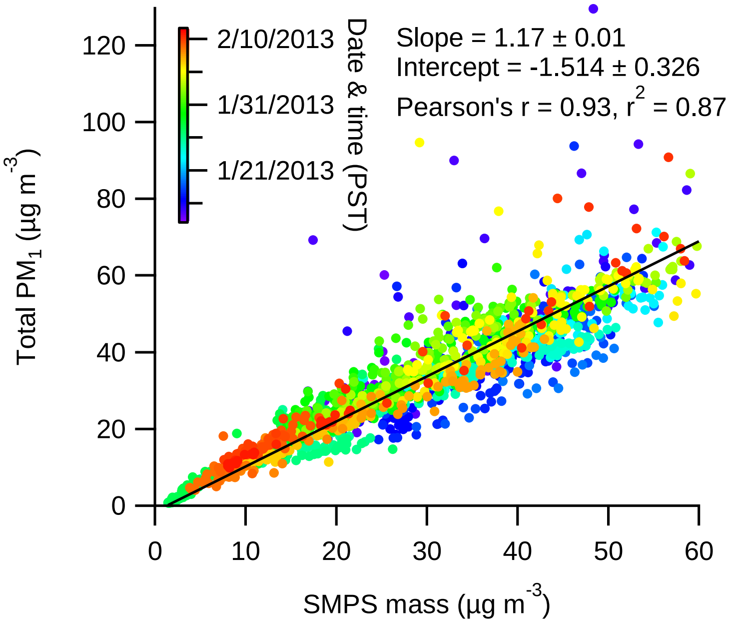
We stated on line 21, P35064 that the calibrations were performed before, during, and after the study. To further clarify, this sentence has been amended in the revised manuscript to include the actual dates of the calibrations as follows: "Ionization efficiency (IE) and particle sizing calibrations were performed following standard protocols (Canagaratna et al., 2007) on January 13, January 19, and February 8."

P35065, L12-14: People who aren't intimately familiar with AMS workings won't understand the point of the statement: "Nitrate was often observed to be an important component of PM1 during this study, yet the campaign average (±1s) CE was 0.5±0.04." Consider rephrasing to make the point clear.

As suggested by the reviewer, these sentences have been rephrased and additional information has been included for clarity in the revised manuscript. Starting on page 35065, line 11, reads: "Although applying a collection efficiency (CE) of 0.5 (default) to whole datasets has been deemed valid for measurements from most ambient environments, several factors, including the relative humidity of the sampling line, ammonium nitrate content, and acidity/neutralization of the sulfate content, have been found to influence the particle phase in the AMS. Consequently, a time- and composition-dependent CE was applied to the data based on the algorithm by Middlebrook et al. (2012). Although nitrate was often observed to be an important component of PM_1 during this study, the campaign average (± 1σ) CE was 0.5 ± 0.04."

P35065, L16-18: "The AMS total mass-based size distribution compares well with the volume size distribution of the SMPS throughout the day (Fig. S2)." is vague. Correlation? Slope? Also wouldn't the relevant comparison be mass vs mass using standard density algorithms to convert SMPS volume to mass?

This statement refers to an image plot showing the size distribution of the AMS mass and SMPS volume as a function of time of day. Thus a visual comparison is made to determine that different aerosol properties i.e. particle size and mass, measured by the AMS compare well with those measured by the SMPS at different times of the day. This follows the initial comparison of the AMS and SMPS measurements as shown in Fig. S1, with strong correlations observed throughout the campaign. However, as suggested by the reviewer, below is a comparison of the mass measured by the AMS and the mass calculated from the SMPS volume, which also shows strong correlation (Pearson's r of 0.93) and slope (1.17). The SMPS mass in the figure below has been calculated using time-varying composition-based densities determined from the AMS measurements. Figure S1 has been updated in the revised supplement accordingly.



P35065, L29: "for comparison to historical measurements"? Again, a non-AMS user will be lost here.

Until the publication of the Canagaratna et al. (2015) paper, all elemental ratios published in the literature were calculated using the Aiken-Ambient method (Aiken et al., 2008). In order to compare the ratios from this study with those from the literature, we report both the Aiken-Ambient and Improved-Ambient ratios. The elemental ratios calculated using the Aiken-Ambient method and Improved-Ambient method are compared and are found to be consistent with the differences between the two methods reported in Canagaratna et al. (2015). The correlation figures are shown below and have been included in the Supplement. For clarity, additional information has been included along with some rephrasing in the revised manuscript, starting at line 27, to read: "This method is an update to the Aiken-Ambient method (Aiken et al., 2008) and is referred to as the Improved-Ambient method. The elemental ratios calculated using both the Aiken-Ambient and Improved-Ambient methods are detailed in Table S1 in the Supplement. The elemental ratios calculated using the Aiken-Ambient method are compared to those calculated using the Improved-Ambient method (Fig. S3) and show high correlation; the slope and r^2 *for O/C are 0.76 and 0.995, for H/C are 0.91 and 0.980, and for OM/OC are 0.92 and 0.988. These* *comparisons are consistent with the average differences for the ratios between the two methods reported in Canagaratna et al. (2015), with increases of 27, 11, and 9 % for O/C, H/C, and OM/OC ratios, respectively."*

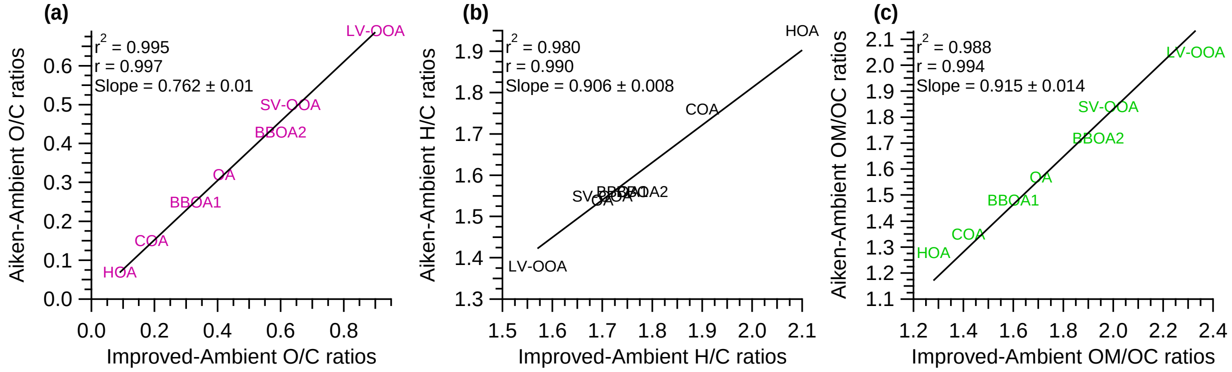


Figure S3. *Comparison of the* ***(a)*** *O/C,* ***(b)*** *H/C, and* ***(c)*** *OM/OC ratios for bulk OA and the six OA factors identified from PMF analysis calculated using the Aiken-Ambient method (Aiken et al., 2008) and the Improved-Ambient method (Canagaratna et al., 2015).*

P35070, L26-29: Can the authors elaborate on why differences in the size distributions is expected to be due to aqueous processing of inorganics. It seems like a broadening could be due to many different factors.

In this study there is evidence that aqueous-phase processing is playing a role in influencing the formation of secondary inorganic species. Ge et al. (2012b) compared the size distributions of aerosol species between foggy periods and no-fog periods in Fresno during 2010 and observed that particles showed a peak at ~500 nm in the accumulation droplet mode, which indicated strong influences from aqueous processes. The peak in particle size is also observed in the current study. However, this is not to say that other factors are not also influencing the observed differences in size distributions. For example, organics are composed of both primary and secondary OA, which are known to have different size distributions, thus the average size distribution for total organic will be somewhere between that for primary and secondary OA, where the secondary OA may also be influenced by aqueous-phase processes.

P35074, L10-12: Not clear if this is referring to the Ge et al. or this study (i.e. are the revised O/C what is noted for all of these studies which are being compared?

As stated on page 35066, lines 1-3, the elemental ratios reported in this paper, both for this study and the other studies that are referred to, are calculated using the updated Canagaratna method. For clarification, these sentences have been rephrased in the revised manuscript and a table has been added to the Supplement as follows: "The COA in this study has an O/C ratio of 0.19, which is lower than the revised O/C ratio for COA in Barcelona (0.27) and New York City (0.23) but is higher than the COA identified in Fresno in 2010 (0.14) (Table S3)."

Table S3. *Comparison of the O/C ratios for COA from various locations calculated using the Improved-Ambient and the Aiken-Ambient methods and the associated references for where the values are reported.*

Location	Improved-Ambient O/C ratio*	Reference	Aiken-Ambient O/C ratio	Reference
Barcelona	0.27	Canagaratna et al. (2015)	0.21	Mohr et al. (2012)
New York City	0.23	Canagaratna et al. (2015)	0.18	Sun et al. (2011)
Fresno	0.14	Canagaratna et al. (2015)	0.11	Ge et al. (2012a)

*Calculated and reported in the Supplement of Canagaratna et al. (2015).

P35076, L26-27: In addition to the references above pointing to a large range of BBOA composition for ambient PMF factors, it would seem useful to reference studies where a range of O/C has been observed for biomass burning source studies, such as the FLAME studies, Carnegie Mellon or PSI groups, etc., since here the supposition is made that the differences may be due to differences in the primary BBOA rather than aged BBOA.

As suggested by the reviewer, additional references have been included in the revised manuscript. Starting on page 35076, line 27, the revised manuscript now reads: "Indeed, a wide range of O/C ratios for primary organic emissions from biomass burning has been observed in various studies (0.15-0.60 e.g. Heringa et al., 2011; Ortega et al., 2013) due to differences in burner type, combustion phases, and fuel types."

P35077, L18-19: "Consequently, adverse health effects associated with biomass burning emissions should be of even greater concern, especially during wintertime." Even greater concern than what?

Biomass burning emissions are already associated with having negative health impacts thus there is already concern surrounding human health and biomass burning. In this study, a strong correlation between BBOA and PAHs was observed; the health impacts from biomass burning could therefore be worse than initially thought due to some PAHs being carcinogenic and mutagenic. To reduce any speculation, this sentence has been rephrased in the revised manuscript to read: "Consequently, adverse health effects associated with biomass burning emissions should be of great concern, especially during wintertime."

P35078, L20-22: "The O/C ratios of both OOA factors are significantly higher than those of the POA factors thus supporting the separation of the factors into primary and secondary OA." This statement doesn't seem like good justification. I.e. the O/C of the SV-OOA is the same as the O/C of BBOA which is a primary factor.

This sentence has been addressed and rephrased in the revised manuscript to read: "The O/C ratio of SV-OOA is higher than HOA and COA and the O/C ratio of LV-OOA is significantly higher than those of the POA factors, consistent with observations that ambient primary OA tend to be less oxidized than secondary OA."

P35079, L10-13: Relevance of correlation if OOA and MSA ions needs more explanation.

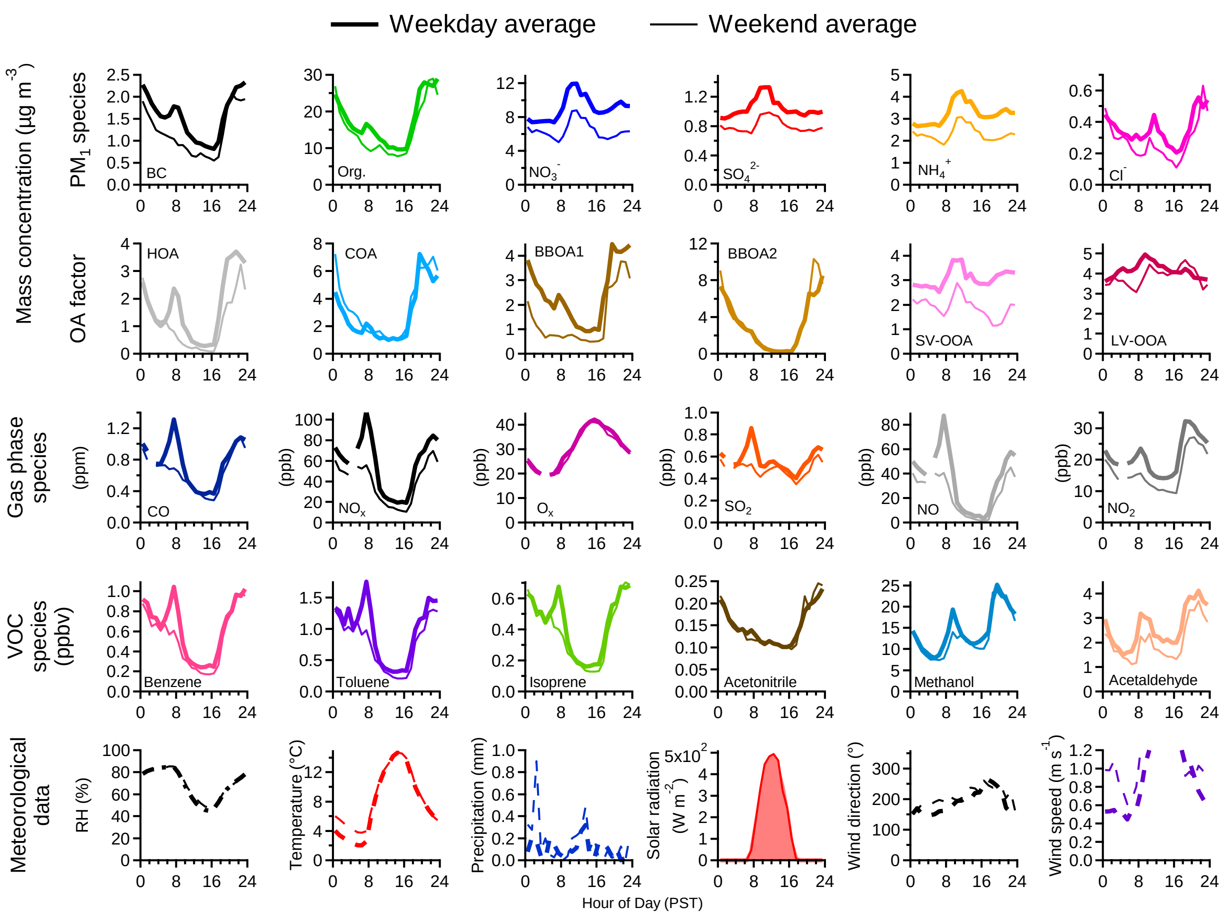
As requested, more explanation has been provided in the revised manuscript. Starting at page 35079, line 10, the revised manuscript now reads: "Methanesulfonic acid (MSA) has been shown to be secondary in previous studies and is mostly the product of dimethyl sulfide (DMS) oxidation (von Glasow and Crutzen, 2004). The AMS has successfully measured MSA in several studies, both over the ocean as well as in urban areas (e.g. Phinney et al., 2006; Zorn et al., 2008; Ge et al., 2012b). Consequently, the secondary nature of SV-OOA and LV-OOA is further supported by strong correlations observed with the AMS spectral ions for MSA (Table 2)."

P35079, L18: Do the authors mean "emissions patterns" (rather than "emissions")

We mean emissions patterns. This has been changed accordingly in the revised manuscript.

P35081, L26-28: O3 may also be larger on weekends due to the O3-formation chemistry being NOx-saturated in which has higher NOx => lower O3. For this reason it would be informative to show the diurnal cycle of Ox = NO2+O3 which helps separate these two effects (NOx-regime O3 chemistry and titration of O3 by NO emission) as has been done in multiple studies (and it seems the authors show for different purposes, 2010/2013 comparison, in Fig 11).

We thank the reviewer for this suggestion. The diurnal profiles for O_x during the weekdays and weekends have replaced those of O_3 in Figures 8 (shown below) and S14 (S15 in the revised supplement). In addition, the text in the revised manuscript has been changed accordingly. Starting on page 35081, line 26 now reads: "Odd oxygen ($O_x = O_3 + NO_2$) is used here to examine the differences between weekdays and weekends rather than O_3 due to the influence of NO_x on O_3 concentrations (Herndon et al., 2008); O_3 is observed to be lower during the weekdays due to titration by NO. The diurnal profile of O_x is therefore comparable between weekday and weekends."



P35082, L12: Too many sig figs.

This has been addressed in the text and associated figure. This sentence in the revised manuscript now reads: "The average PM_1 concentration was higher during the first event than the second event (44 μg m^{-3} compared to 36 μg m^{-3}) (Fig. 2h and j) and the average compositions of PM_1 are also fairly different."

P35085, L12-14: "At around 17:00 PST the sun sets, the boundary layer starts to collapse, and any pollutants that mixed aloft during the day would be decoupled from the surface." Please provide the reasoning for choosing that time. A rise in CO along would probably not be precise since the rise appears to be coincident with the evening rush hour. Perhaps the weekend CO profile supports this statement? Or something else the authors have in mind?

*This time was chosen for several reasons. Firstly, the diurnal profile of NO_2*solar radiation as shown in Figure 11 reaches zero between 17:00 and 18:00 PST, indicating that the sun has set by this time. Secondly, previous studies in this region such as that by Watson and Chow (2002) suggest that the sun sets between 17:00 and 18:00 PST. Finally, as also suggested by the reviewer, the onset of the evening increase in CO concentrations at the weekend occurs at approximately 17:00 PST. However, the CO profile is very similar in the evenings during the weekdays and weekends indicating that boundary layer dynamics are more important in influencing CO concentrations than rush hour emissions in the* *evening. Additional information has been included in the revised manuscript. Starting at page 35085, line 12, the revised manuscript now reads: "The CO profile is very similar in the evenings during the weekdays and weekend (Fig. 8) indicating that boundary layer dynamics are more important in influencing CO concentrations than rush hour emissions in the evening. Thus at around 17:00 PST the sun sets, the boundary layer starts to collapse, and pollutants that mixed aloft during the day would be decoupled from the surface (Pusede et al., 2016)."*

P35085, L16-17: "Ox is used here to indicate HNO3 production, although N2O5 chemistry can also influence the formation of HNO3": Please elaborate. It is not clear how Ox would be a direct surrogate for HNO3.

We are not suggesting that O_x *is a direct surrogate for* HNO_3 *but rather it is being used in this analysis to indicate whether* HNO_3 *production is likely and how it compares between 2010 and 2013. The reactions for this, as reported in Seinfeld and Pandis (2006), are:*

$$NO_2 + O_3 \rightarrow NO_3 + O_2$$

$$NO_3 + NO_2 + M \rightarrow N_2O_5 + M$$

$$N_2O_5 + H_2O \rightarrow 2HNO_3$$

FIGURES:

Fig. 1b: Can't read features like "FWY-41" on the map. Perhaps it is just the rendering of the pdf, but best to be able to identify features that are pointed out in the text.

The Figure has been amended accordingly in the revised manuscript.

Fig 2e: It doesn't explain in the legends or caption what the gray shading is on this plot?

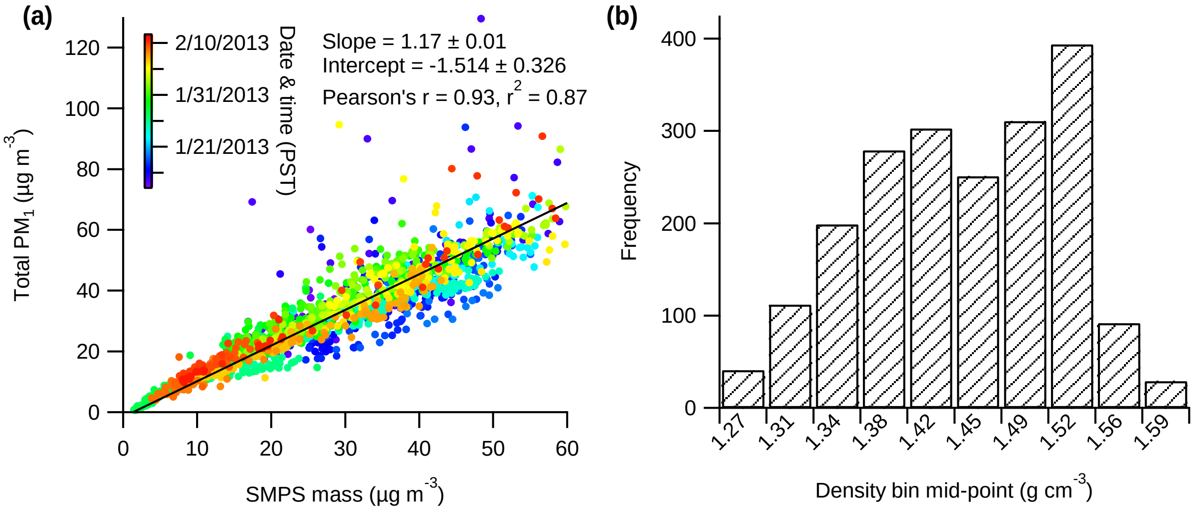
*The caption for Fig. 2e has been amended in the revised manuscript to include the explanation for the gray shading as follows: "**(e)** time series of total* PM_1 *and SMPS mass concentrations where SMPS mass was calculated using a time-varying density based on measured particle composition (see Fig. S1b). Also shown are the 24-hour average National Ambient Air Quality Standard for* $PM_{2.5}$ *(35 µg* m^{-3}*) and the calculated average daily* $PM_{2.5}$ *concentrations for comparison. Persistent exceedances of this standard characterize the two pollution periods highlighted by the gray shading (14-23 January and 29 January–5 February);"*

Fig 2e: For SMPS, it would seem more appropriate to use a time-varying density as clearly there appears to be a substantial variability in density, in part seen by the relative change in inorganic/organic contributions in Fig 2.

As suggested by the reviewer, the time series of SMPS mass has been calculated using a time-dependent density and the figure has been updated accordingly in the revised manuscript.

Fig. S1: References for PMF factor densities?

References have now been given for the densities of all species used in this calculation. The density of OA was calculated for this study based on the method reported in Kuwata et al. (2012) using the O/C and H/C ratios for bulk OA. Figure S1 and its caption have been updated in the revised *supplement as follows, where Figure S1a has been changed from a scatter plot of AMS mass versus SMPS volume to AMS mass versus SMPS mass in response to another comment:*



***"Figure S1. (a)** Scatter plot of the total PM$_1$ mass (NR-PM$_1$ plus BC) versus SMPS mass, where the NR-PM$_1$ has been corrected using a time- and composition-dependent collection efficiency (Middlebrook et al., 2012). The SMPS mass was calculated using a time-varying composition dependent density from the AMS. The density was calculated based on PM$_1$ composition, which consists of ~34% ammonium nitrate (density = 1.72 g cm^{-3}), ~4.3% ammonium sulfate (density = 1.77 g cm^{-3}), ~1.6% ammonium chloride (density = 1.52 g cm^{-3}), ~55% OA (density = 1.18 g cm^{-3}), and ~4.8% BC (density = 1.77 g cm^{-3}), averaging 1.44 g cm^{-3}. The densities for ammonium nitrate and ammonium sulfate are from Cross et al. (2007), for ammonium chloride the value is from Haynes (2014), the OA density was calculated using the method reported in Kuwata et al. (2012) based on the O/C and H/C ratios for bulk OA, and the BC density is from Cross et al. (2007); Zhang et al. (2015); **(b)** histogram of particle density calculated based on PM$_1$ composition."*

Fig S6: It would be useful to write the species somewhere on this plot (e.g. on the blue areas at small sizes in white or yellow or next to the panel letter on top).

Fig. S6 has been updated in the revised Supplement to include the species for each individual plot. This figure is now Fig. S7 in the revised supplement as additional figures have been included in response to other comments.

Responses to Anonymous Referee #2

General comments

This is an extremely well written, appropriately cited manuscript that focuses on the analysis of particulate matter data from the San Joaquin Valley during the DISCOVERAQ campaign in January/February 2013. It is very thorough with regard to the information included and with regard to the analyses performed. The figures show an incredible amount of information, and as a result can be difficult to read as too much is crammed into individual figures. All that being said, I would say that the analyses performed are now considered somewhat standard for the AMS community. In no way do I mean to minimize the work; it is difficult and time consuming to perform analyses at this level. I only state this because the generalizable insight gained is limited. I have had manuscripts criticized and rejected for similar reasons. I personally would err on the side of publication (assuming specific comments below are addressed) as it is important to have such high quality data in print, but I would defer to the editor.

Technical/significant comments

Page 35063, line 6. “cold” and “relatively dry” are generic. Please provide specific information (data).

To support these descriptions of the weather during the campaign, data has now been included in this sentence as requested in the revised manuscript. The sentence now reads: “During this time, the weather was cold (average temperature of 7.9 °C) and relatively dry (average RH of 69 %) with frequent sunshine.”

Page 35065, line 7. Was measured CO2 used to do this? Source of that data? Also, does the isotope ratio vary from location to location for N2 in the atmosphere?

Unfortunately gas-phase CO_2 measurements are not available from this site during the campaign. As mentioned in line 4, the modifications to the fragmentation table were based on data from filtered air periods. Filtered air measurements are particle free thus any signals detected by the AMS are those of background gas-phase signals. These signals are removed from the data, resulting in measurements of just particle-phase species. Note that due to high organic aerosol loading during this study, gas-phase contribution represented a minor fraction of the total CO_2^+ signal and using a constant background CO_2^+ subtraction had little influence on the determination of org-CO_2^+ signals.

The nitrogen isotopic ratio for N_2 in the atmosphere should be nearly constant. However, the ratio of $^{15}N^{14}N^+$ (m/z = 29) to $^{14}N^+$ (m/z = 28) determined by the HR-ToF-AMS is usually biased low compared to the natural isotopic ratio for $^{15}N/^{14}N$ due to instrument settings. It is therefore a common practice to determine this ratio using filtered air during a field study.

Additional information has been included in the revised manuscript. Starting at page 35065, line 8 now reads: “For improved oxygen-to-carbon (O/C) estimations, there is a need to perform a time-dependent CO2+ subtraction (Collier and Zhang, 2013), however, due to high organic aerosol loading during this study, gas-phase contribution represented a minor fraction of the total CO2+ signal and using a constant background CO2+ subtraction had little influence on the determination of org-CO2+ signals.”

Page 35066. I would recommend considering a reorganization of the text to discuss PMF prior to the elemental analysis. When I referred to Table S1 (line 3), it brought in the PMF factors even though they had not been defined or discussed yet. This suggestion would help improve the flow of the paper for those that might be less familiar with AMS terminology. Note that on line 3, Table S2 does not actually contain the information stated at this point (its reference later in the manuscript is correct).

We thank the reviewer for the suggestion to reorder the text. However we feel that the current order is more appropriate and in a logical ordering of data analysis given that elemental ratios can be calculated for the total organic fraction irrespective of whether PMF is performed or not. The average elemental ratios for bulk OA during this study are included in Table S1. In this study, PMF is applied to apportion the total organic aerosol fraction into its components where elemental ratios are calculated for each identified source (factor) and reported in Fig. S4, which is Fig. S5 in the revised supplement. In addition, the differences in the degree of oxidation (as indicated by the O/C ratio) between potential sources and those identified in previous studies are used to help determine the appropriate number of factors in this study (lines 14-17). Consequently, elemental ratios need to be discussed before the PMF section.

The reference to Table S2 in line 3 is to the table in Canagaratna et al. (2015), not the table in this manuscript. To improve the clarity, this sentence has been amended to read: "Unless otherwise indicated, the O/C, H/C, and OM/OC ratios stated in this paper from other studies have been calculated using the updated elemental analysis method and are detailed in the Supplement of Canagaratna et al. (2015) (Table S1 and S2)."

Page 35067. This is a lot of detail for non-AMS users. It is fine to leave it in, but if that is done, I would recommend defining some of the parameters - for example fpeak, Q, etc.

As recommended by the reviewer some of the PMF parameters have been defined in the revised manuscript. This information has been included starting at page 35067, line 4, to read "Within the PMF analysis toolkit, there are several diagnostics that can be used to aid the choosing of the best modeled number of factors including Q and f_{Peak}. Q is the quality of fit parameter and the ratio of the expected Q (Q/Q_{exp}) indicates how well the model fit the data; as the number of factors in a solution increase, the degrees of freedom increase and Q/Q_{exp} decreases close to 1 as more data are able to be fit. The f_{Peak} parameter is used to demonstrate the variation of the solutions and can indicate the rotational stability of the solution sets. Negative f_{Peak} values result in variations in the time series and positive values result in variations in the mass spectra of the solutions. However, the solution set is most likely to be physically meaningful when f_{Peak} is zero (Paatero et al., 2002). In this study, the rotational stability of each of the solution sets were explored through the f_{Peak} parameter from -1 to 1,…"

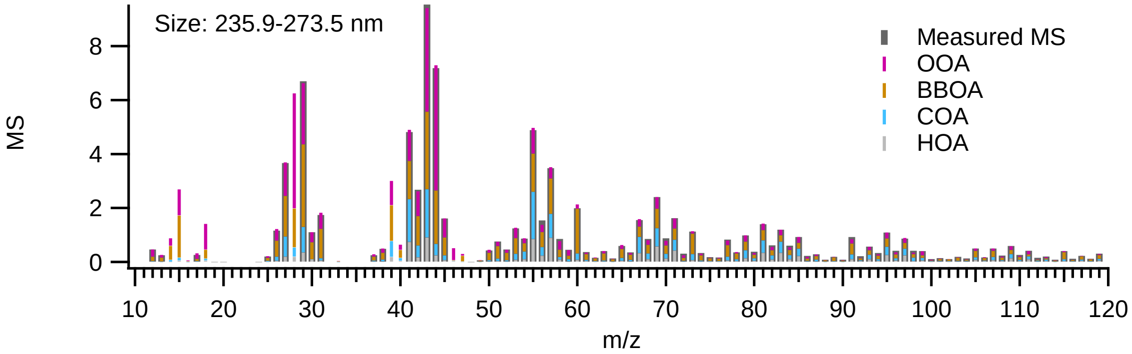
Page 35067, line 21. What is the basis for saying that "Factor 2 appears to be a combination of the other two factors." Visual inspection?

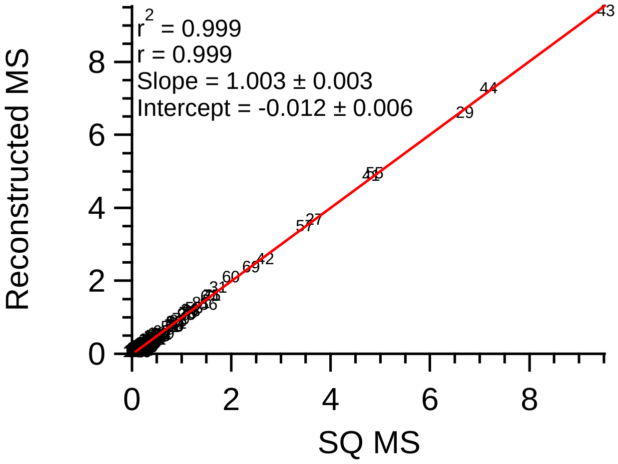
Visual inspection does indicate that the mass spectrum of Factor 2 of the 7-factor solution is a combination of the mass spectra of Factors 1 and 4. In particular, the key peak in Factor 1 is at m/z 44, composed predominantly of CO_2^+, and the key peak in Factor 4 is at m/z 43, composed predominantly of $C_2H_3O^+$, and Factor 2 has key peaks at m/z's 43 and 44 with similar compositions.

Page 35068, line 10. What is the validity of assuming the spectral profile of each factor is constant within each size bin? I can imagine, for example, that that of biomass burning aerosol varies depending on the type of particle (larger more ashlike versus smaller formed from condensation of gases upon cooling).

As PMF is used to separate the organic aerosol fraction into different components according to composition/processes, we assume the composition of each factor, as represented by their respective mass spectra, is the same in each size bin. In other words, the type of particle has already been determined via PMF analysis thus estimating the sizes of each of the particle types is a next step in the analysis.

To validate this assumption the total reconstructed mass spectrum of the four OA factors used in this analysis (the two BBOA factors were summed and the two SOA factors were summed) for each size bin were compared to the measured mass spectrum for that size bin. The r^2 *for the fit ranged from 0.853 to 0.999 and Pearson's r ranged from 0.923 to 0.999 where the lower values correspond to the fits from the smallest size bin (40-46.4 nm). An example of this information is shown below; MS stands for mass spectrum, and SQ MS denotes the summed unit mass resolution mass spectrum. Consequently, the assumption that the spectral profile of each factor is constant within each size bin.*



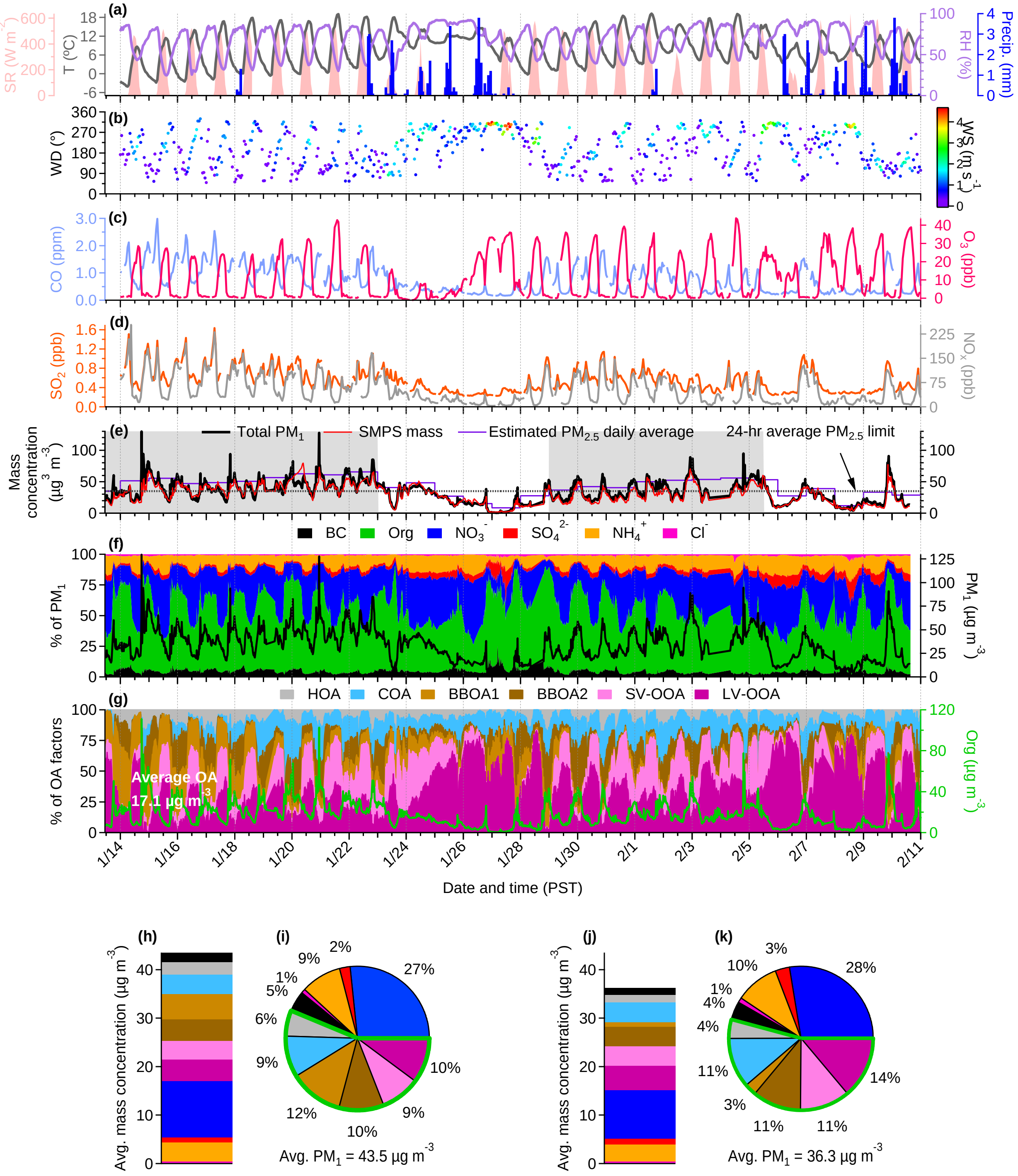


Section 2.3.4 Are there NH3 and HNO3 data available to help analyze/understand the KAN calculations performed and shown later in the manuscript?

As mentioned on page 35084, line 24-25, there are HNO_3 *and* NH_3 *data available for the 2013 campaign, however the measurements are only approximately every 7 hours. Consequently, the time-resolution was deemed too low to investigate the variations in ammonium nitrate concentrations and the* K_{AN} *calculations. However, as detailed on page 35084, line 21-24, it is likely that most* HNO_3 *formed has likely partitioned to the particle-phase in this study as indicated by the estimate that approximately 89% of the total nitrate is in the particle phase based on the* HNO_3 *and particle-phase nitrate measurements.*

Page 35069, line 18. If statements are made about exceedances of the NAAQS, I think it is important that the basis for those statements are included. For example, use the 25% factor to estimate PM2.5 and take a 24-hour average. Compare those numbers to the standard. This can also be done on Figure 1 (the NAAQS is shown, but estimated 24-hour average PM2.5 data are not, which can be misleading).

The calculations have been performed and the data included in Figure 2, as suggested by the reviewer. The figure and figure caption have been updated in the revised manuscript. The updated figure looks as follows:



Page 35070, line 10 (as well as on pages 35085 and 35086). In this and other locations in the manuscript, the importance of boundary layer dynamics is discussed. This is suppositional and based on temporal profiles of pollutants. However, some effort should be made to consider what the BL heights are, as well as their temporal dynamics. Were any soundings made? LIDAR?

Unfortunately the only measurements for boundary layer were made on board the aircraft rather than in situ. In addition, when the aircraft passed over the site in Fresno, it was approximately 10:00-11:00 am thus the boundary layer dynamics referred to in this manuscript (e.g. morning break-up and evening collapse) have not been captured due to the timings of the aircraft measurements. Nevertheless, Pusede et al. (2016) estimated daytime boundary layer heights during DISCOVER-AQ as ranging between 300 to 700 m based on aerosol backscatter estimates by a micro-pulse lidar (MPL) supplemented with a wide-field receiver system as well as various other measurements including NO and RH from the P-3B aircraft.

Page 35072, line 1. The authors state that N appears to be associated with SOA. Are sufficient data available to also perform the Fry/Farmer analyses that apportion AMS nitrate to inorganic and organic forms?

This statement refers to the N associated with the organic aerosol fraction i.e. N-containing organic ions with nominal formula of $C_xH_yO_zN_p^+$ *(*$x > 0; y \geq 0; z \geq 0; p \geq 1$*), rather than nitrate. We have examined the correlation between* NO^+ *and* NO_2^+ *signals and found that these two ions are highly correlated (*$r^2 = 0.967$*) and that the* NO^+/NO_2^+ *ratios for ambient aerosol were very similar to the ratio of the pure ammonium nitrate from the calibrations. In addition, the nitrate concentration measured by the AMS correlates very well with that measured by the PILS (Parworth et al., In preparation). We therefore conclude that the nitrate concentrations measured by the AMS were for inorganic nitrate, mostly ammonium nitrate.*

Page 35078. Were any filters collected at this site during this time, as is often done during such large campaigns? Speciation of organic matter, specifically regarding levoglucosan, its isomers, and other tracers of biomass burning, would be incredibly helpful in the discussion of the differences between the biomass burning factors.

Unfortunately, we did not collect filter samples during this study.

Page 35078, line 18. There is so much in the SI that it would be appropriate to include an example of the thermodenuder data to which the paper refers.

Analysis of the thermodenuder (TD) data is still ongoing thus we are unable to present it here. However, we have examined the average mass spectrum of organic aerosols at different TD temperatures. We have performed a linear decomposition of the bulk organic mass spectrum according to the different factors at each temperature and the preliminary results indicate that LV-OOA is of lower volatility as it contributes more to the mass spectrum at higher TD temperatures. We intend to do a more thorough analysis of the TD data and report the results in a future publication.

Page 35080, line 7. Based on recent work from the SOAS campaign as well as from Carlton et al., RH is potentially not the best metric to evaluate the likelihood of aqueousphase processing. Methods using ISORROPIA (corrected using a kappa for organics) can estimate aerosol liquid water content. It is suggested to perform such calculations.

We are currently performing such analysis using the PILS data and plan to discuss the results in a separate paper (Parworth et al., In preparation) that focuses on the PILS observations.

Section 3.4.2 I find the comparison between the two winters relatively less interesting. The meteorology is clearly different, so one would expect the PM to be as well. That being said, are there other aspects of the data set that can be used to explore? For example, the estimated size distributions of the factors; these are not often discussed in the AMS community. Can more information be gleaned from this?

As suggested by the reviewer, comparisons were made between the estimated size distributions of the OA factors as well as nitrate and sulfate from 2013 and 2010 (Ge et al., 2012b); the particle sizes in 2013 are narrower and larger than 2010. The differences appear to be consistent with overall more aged aerosol in 2013, which is likely related to the more stagnant meteorological conditions in 2013. The comparison of the size distribution has been added in the revised manuscript, with the associated figures in the supplement (as shown below). Starting at page 35084, line 4, the revised manuscript now reads: "The estimated size distributions of the OA factors, as well as nitrate and sulfate, were compared between 2013 and 2010 (Ge et al., 2012b); the particle sizes are observed to be narrower and larger in 2013 than in 2010 (Fig. S19). These differences appear to be consistent with overall more aged aerosol in 2013, which is likely related to the more stagnant meteorological conditions."

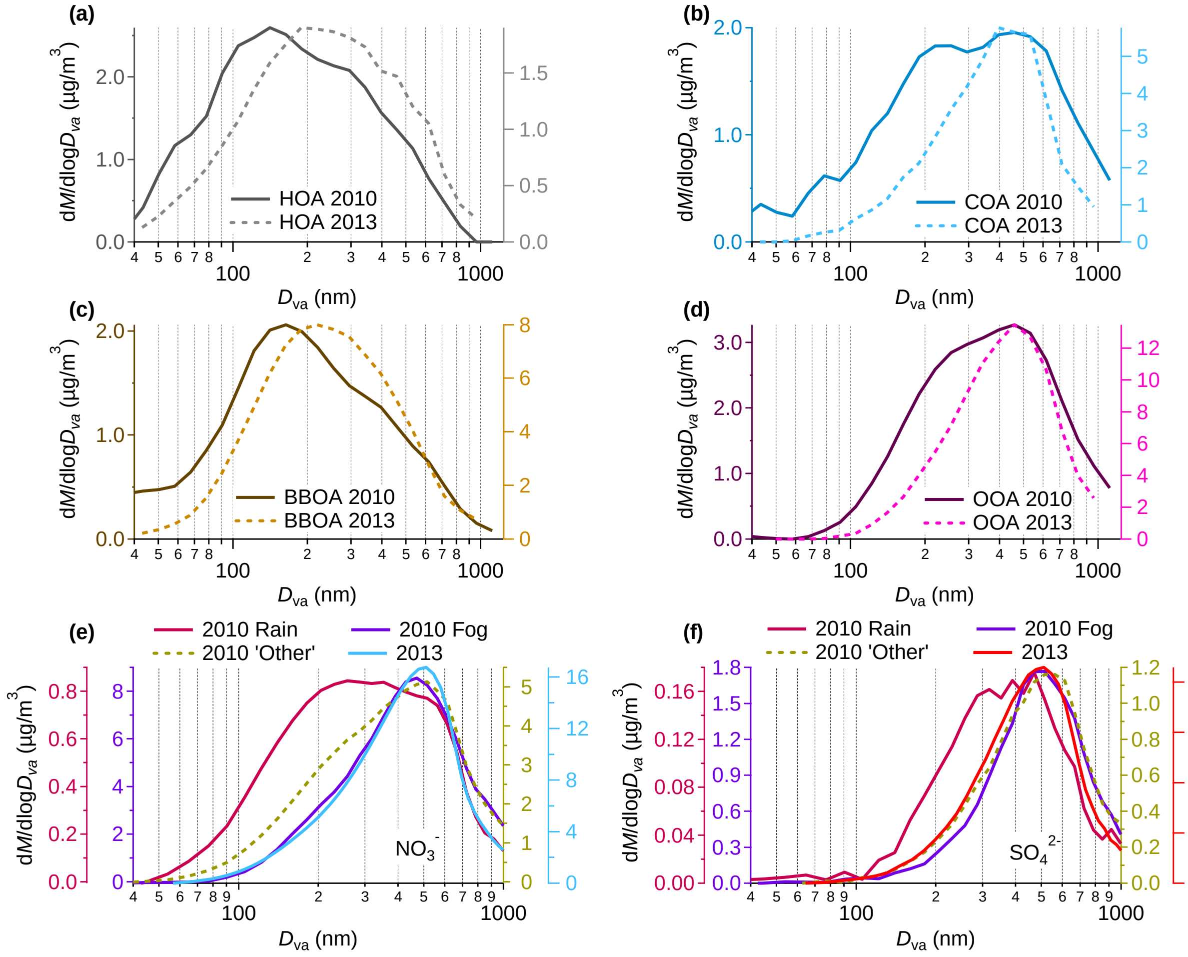


Figure S19. *Comparisons of the average size distributions between 2013 and 2010 (Ge et al., 2012b) for the estimated size distributions of the OA factors* ***(a)*** *HOA,* ***(b)*** *COA,* ***(c)*** *BBOA, and* ***(d)*** *OOA.* ***(e)*** *and* ***(f)*** *show the average size distribution of nitrate and sulfate, respectively, from 2013 compared with the average size distribution of the same species from 2010 during different meteorological conditions, defined as rain, fog, and 'other' (see Ge et al. (2012b) for further details).*

Page 35085, line 21. The potential influence of N2O5 on nitrate is mentioned, but no effort is made to quantify it. Is this possible, even to first order?

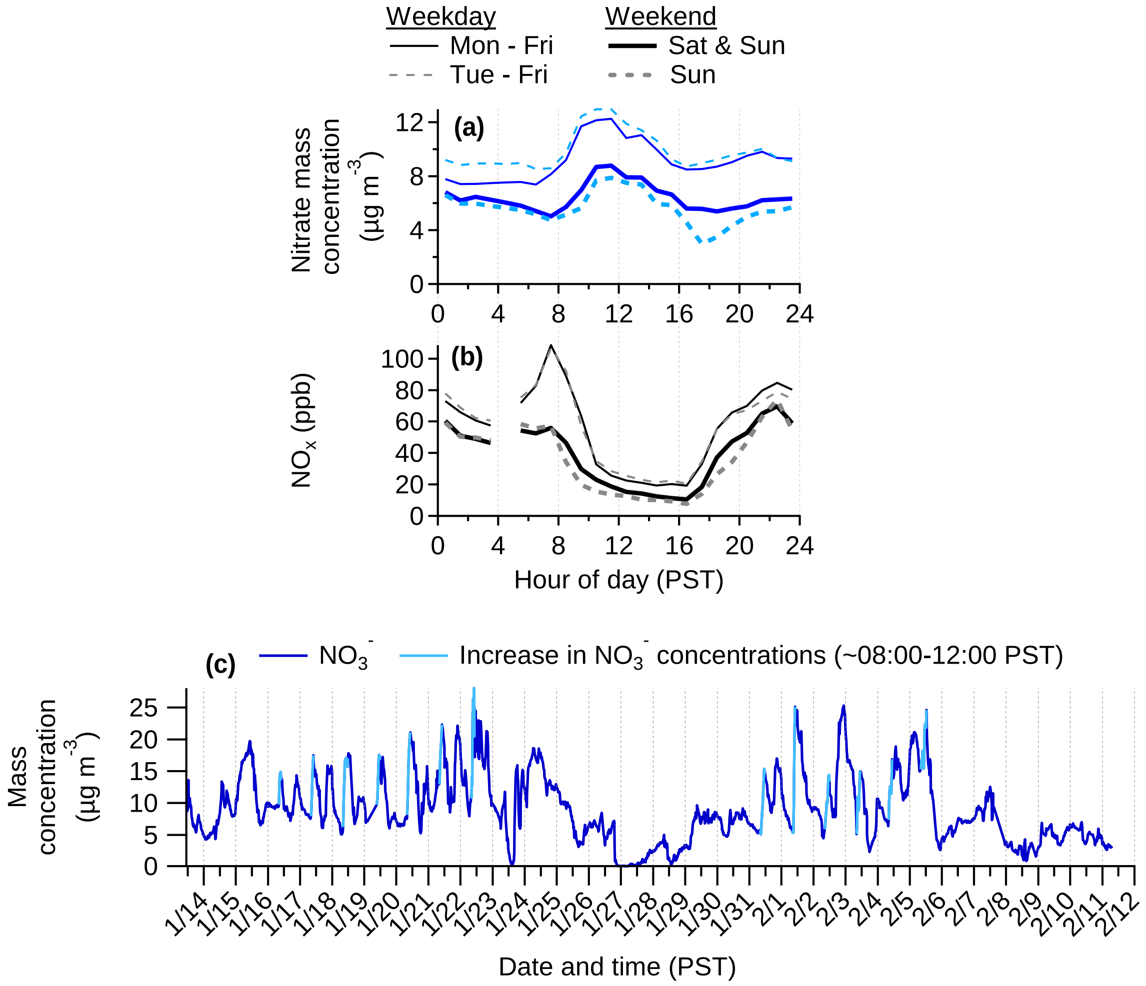
These calculations and associated analyses are being performed using ground site and aircraft measurements; the results will be discussed in a separate publication (Prabhakar et al., in preparation). To a first order, it is indicated that a predominant fraction of the available NO_x *in the residual layer is converted to* HNO_3 *via the* N_2O_5 *pathway overnight. It should be noted that* $ClNO_2$ *can also be formed from heterogeneous reactions of* N_2O_5 *which would lead to lower* HNO_3 *production. However, the importance of the* $ClNO_2$ *pathway varies with day and on average,* HNO_3 *production from* N_2O_5 *is the dominant pathway.*

Figure 10. Is the estimate of BC in 2010 relative to PM1 valid? Would a ratio to POA be better? This might not make a significant difference in the data, but it would be more defensible, I believe.

As suggested by the reviewer, the ratio of BC to POA in 2013 has now been used to estimate the concentration of BC in 2010. This has not made any significant difference to the results as the changes were negligible, although the data and figure have been updated accordingly in the revised manuscript.

Figure 13c. "Morning increases" are inadequately described.

*Assuming the reviewer is referring to Figure S13c in the supplement, both the legend in the plot and the figure caption have been amended in the revised Supplement accordingly. The figure number has been changed to S14 as additional figures have now been included in response to other comments. The figure caption reads: "****Figure S14.*** *Average diurnal mass concentrations when different definitions of weekdays and weekends are used for* ***(a)*** *nitrate and* ***(b)*** NO_x*, a gaseous precursor of particulate nitrate.* ***(c)*** *Time series of nitrate highlighted with the occasions when a rapid increase in concentration during the morning (between approximately 08:00 and 12:00 PST) is observed."*



Minor comments

Table 1. Would be appropriate to include standard deviations in last column as well. Same with first two rows of Table 3.

As suggested by the reviewer, Table 1 has been amended accordingly in the revised manuscript and the top two rows in Table 3 have also been amended.

Figure 1. Are CRD-PAS, PEX, and APS even used here? If not, could remove for simplicity.

The data from the instruments mention in the reviewer's comment are not used in the analysis presented in this manuscript. However, the data are used in other analyses and reported in published manuscripts (e.g. Zhang et al., 2016) thus we feel that it is useful to keep these instruments in the figure for reference.

Figure 9. The x axis label needs to be corrected (ug/m3, not C)

The x-axis label has been corrected accordingly.

Table S2. Does the reference to Ge et al. refer to a, b, or both?

In the Supplement, only one of the Ge et al., 2012 manuscripts is referred to. As such, no changes have been made to the citation and corresponding reference in the Supplement. For clarification for the reviewer, the Ge et al., 2012 in the Supplement is the same as Ge et al., 2012a in the main manuscript.

Figure S5. Are the data presented average values? Or for a specific subset of the data? Please specify in the caption.

*As stated in the figure caption, the data presented are for each size bin and are for 40-1200 nm in figure S5a, and the data for size bins covering 70-1000nm are shown in S5b, c, and d. Other than the two BBOA and the two OOA factors being grouped to result in four main OA factors, the data presented are for each size bin covering the whole study. To clarify this, the caption has been amended, and the figure number updated, in the revised supplement to read: "**Figure S6.** Summary of key diagnostics from the fitting of the derived size distributions of the four main OA factors from the whole measurement campaign: **(a)**…"*

Figure S11. The caption should refer to being colored by date, not time of day.

*The text for this figure caption has been amended accordingly and the figure number amended due to additional figures now included. The text in the revised supplement now reads: "**Figure S12.** Triangle plots of **(a)** f_{44} vs. f_{60} and **(b)** f_{44} vs. f_{43} for the six OA factors and all measured OA data (dots), colored by date."*

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