We thank the reviewers for their comments. Our specific response can be found below. The reviewers' comments are in italics and changes made to the manuscript are in quotation marks. All changes made are minor and do not affect the conclusions in the manuscript.

Response to Reviewer 1

1. The authors state (P10484, L14-16) that Budisulistiorini et al. used factor analysis to identify various OA sources, however it is not clear how this specific study shows "the versatility of PMF analysis in OA source apportionment" as the authors state. Please clarify.

Response: We agree with reviewer that the phrase "indicating the versatility of PMF analysis in OA source apportionment" is overstated and have deleted the phrase in the revised manuscript.

2. Why were the organic fractions of NO+ and NO2+ not included in the determination of the elemental ratios, particularly when organic nitrates contributed significantly to the total OA? Would inclusion of the organic fraction of these ions make a significant difference in the O:C or OS values?

Response: We didn't include NO^+ and NO_2^+ in the calculation of elemental ratios for the following two reasons. Firstly, this allows for a more direct comparison with literature values, which typically do not include NO^+ and NO_2^+. Secondly, including NO^+ and NO_2^+ does not affect the O:C or OS values significantly. Take CTR_June as example, where almost all nitrate is estimated to arise from organic nitrate. Including NO^+ and NO_2^+ only increases the O:C value from 0.75 to 0.77.

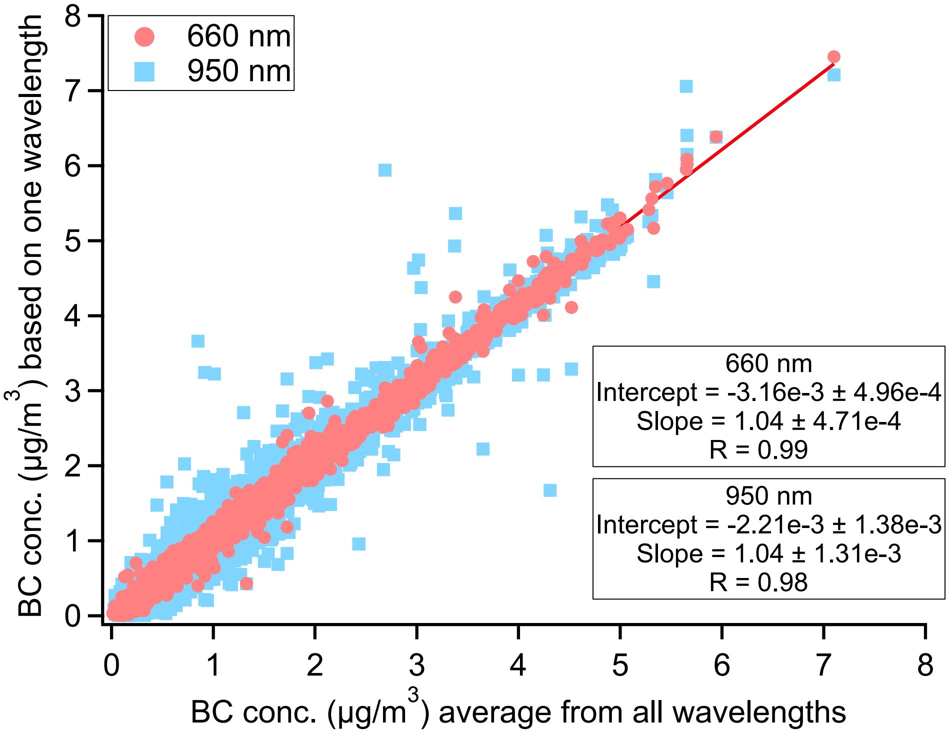
3. The authors give the RIE values used when analyzing ACSM data (P10488-10489), but not for the AMS data. What values were used for the AMS data?

Response: Default RIE values are used for the AMS data. For ammonium, the average RIE determined from IE calibrations by using ammonium nitrate is 4, which is the default value. We have added the following sentence in the revised manuscript.

"Default RIE values were used for the HR-ToF-AMS data."

4. For the Aethalometer data, black carbon concentrations were determined by averaging the signal determined at all seven wavelengths measured by the instrument. However, absorption at the shorter wavelengths can be enhanced by the presence of organics. Biomass burning in particular emits organics which absorb at the shorter wavelengths. Why was the black carbon concentration determined using an average of all seven wavelengths, as opposed to just using the longer wavelength(s), which theoretically have fewer interferences from non-black carbon material?

Response: The BC concentration calculated using an average of all seven wavelengths is very close to that using just one longer wavelength. Take the GT_Aug site for example, in the figure below, we compare the BC concentration based on 660nm or 950nm with the BC concentration based on average of all wavelengths. The difference is only 4%.



To make things clearer, we will report the BC concentration based on 660nm absorption in the revised manuscript as the reviewer suggested. 660nm is chosen because it is the closest to the MAAP wavelength (i.e., 670 nm), which was deployed at three sites in this study.

5. In addition to isoprene and β-pinene, Bruns et al., 2010 also give Rorg for several other biogenics (i.e., α-pinene, limonene and 3-carene), which have larger Rorg values than for isoprene and β-pinene (although the α-pinene Rorg is only slightly larger than that of β-pinene). Why weren't these other biogenic considered in the determination of the upper and lower values of Rorg in this study? This should be noted in the manuscript.

Response: As stated in the manuscript, "we select organic nitrates formed from isoprene and β-pinene oxidations as representative because of their large abundance in the southeastern US, potential to produce organic nitrates, and that they cover a wide range of R_{ON}/R_{AN} values". The organic nitrate formed from limonene, 3-carene, and α-pinene are not considered due to the following reasons. Firstly, the concentrations of limonene and 3-carene in the southeastern US are much lower than that of isoprene and β-pinene. For example, campaign-averaged nighttime concentration of limonene in CTR_June was only 0.05ppb, while it was 0.31 ppb for β-pinene (Xu et al., 2015). Secondly, while the concentration of α-pinene is similar to that of β-pinene, the SOA yield and organic nitrate of α-pinene is much lower than those of β-pinene (Berndt and Boge, 1997; Boyd et al., 2015; Griffin et al., 1999). Thirdly, as reported in Bruns et al. (2010), the NO^+/NO_2^+ ratios of organic nitrate from α-pinene, limonene, and 3-carene have much larger uncertainties than those of organic nitrate from isoprene and β-pinene. For example, the average + 2 standard deviations are 11 ± 8, 15 ± 8, and 14 ± 12 for α-pinene, limonene, and 3-carene derived organic nitrates, and the values are 10 ± 2 and 5.0 ± 0.7 for β-pinene and isoprene derived organic nitrates.

We have added the following sentence in the revised manuscript.

"The organic nitrates derived from other biogenic VOCs (i.e., α-pinene, limonene, 3-carene, etc) are not considered due to either their lower ambient concentrations in the SE US or lower organic nitrate yields compared to isoprene and β-pinene (Xu et al., 2015)."

6. I have several comments on Figures 3 and 6. Firstly, the authors state that the OA diurnals reach a daily maximum in the early morning and evening (except for YRK_July). Are the authors describing two maximums during the day (one in the early morning and one in the evening), or *one maximum that occurs either during the evening or the early morning? If it is the latter, the peak is in the night/early morning for panel (a, c, f, g, h) in Figure 3. The peak is only in the evening for CTR_June with BLH adjustment. In the conclusions, the authors state that the OA diurnal profiles peak at night in the winter datasets. The description of the diurnal patterns should be clarified. Secondly, there are no error bars given on the diurnals in Figure 3 (or Figure 6), making it difficult to determine if the diurnal trends are significant. For example, are the differences in the GT_Aug OA diurnal, which has little variability, significant? Can error bars be added to the diurnals? Finally, should the units on panel (d) in Figure 3 still be μg m-3, as the concentrations have been multiplied by the planetary boundary layer height?*

Response: Firstly, we are describing that for the summer datasets, the OA shows a slight increase at night and reaches a daily maximum either during the evening or the early morning. The key point of OA diurnal variation is that the OA diurnal trend in summer is relatively flat. The lack of a prominent daytime increase in the OA in summer would at first appear to discount the role of photochemistry-driven secondary OA formation. We interpret the flat OA diurnal trend in summer based on the changes in planetary boundary layer height and changes in contributions to total OA from various sources. When taking the changes in BLH into account, there is net OA production during the day in summer. We have rephrased the description of OA diurnal variation.

"In terms of diurnal variation, the OA diurnal trend is relatively flat in summer and peaks at night in winter (Fig. 3). The diurnal variation of OA is largely influenced by the changes in planetary boundary layer height and changes in contributions to total OA from various sources, which will be discussed in detail in section 4.4.1."

Secondly, we have added the error bars in Figs. 3 and 6 as the reviewer suggested.

Finally, on panel (d) in Fig. 3, the unit of y-axis is indeed μg m^{-2}, instead of μg m^{-3}. The confusion is caused by that the axis labels are not readable in the ACPD version. Once the manuscript gets accepted by ACP, we will request that figure 3 takes up a whole page and we believe the figure will be readable then.

7. In section 4.1.2, the authors describe the COA factor as having a small peak at lunch time and a larger peak at dinner time in all datasets where a COA factor was identified. However, I do not *see a lunch time peak in the JST_Nov data in Figure 6f. Why is there no peak in the COA diurnal during lunch time in JST_Nov?*

Response: For the three out of four datasets where a COA factor is resolved, the COA exhibits a small peak at lunch time and a larger peak at dinner time. The only exception is JST_Nov dataset, where COA starts decreasing since ~6am and reaches daily minimum ~3pm. Although the reason for the lack of a lunch-time peak in JST_Nov is unclear at this moment, the mass spectrum of COA factor in JST_Nov is similar to that of other sites in the current study and it is also similar to the COA mass spectra in the literature, which supports the identification of COA for JST_Nov dataset.

We have amended the text in the revised manuscript for clarity.

"Another feature of COA is its unique diurnal trend. For three out of four datasets (except JST_Nov) where a COA factor is identified, the COA factor exhibits a small peak at lunch time and a large peak at dinner time (Fig. 5)."

8. When discussing the Isoprene-OA factor, the authors state that fC5H6O+ is higher at rural sites than urban sites, which could be explained by advection from rural sites to urban areas, during which time compounds giving rise to C5H6O+ are further transformed. However, the authors conclude based on other evidence that advection is likely not significant. What then is the explanation for the lower fC5H6O+ in urban sites? Could this be an indicator of mixing of the PMF factors in the urban datasets?

Response: In this study, we do not have definite explanations regarding why f_{C5H6O+} is higher at the rural sites than urban sites. Factor mixing in the PMF analysis and the influence of seasonality (as proposed by Reviewer#3 comment#8) are possible explanations. We have added the following discussions in the revised manuscript.

"Another possibility for the lower f_{C5H6O+} at the urban sites is that Isoprene-OA factor from the urban sites may contain isoprene SOA produced via other pathways, in addition to the IEPOX uptake pathway. Isoprene SOA formed via RO_2+NO pathway only has a negligible signal at $C_5H_6O^+$ (Kroll et al., 2006; Xu et al., 2014), so that the mixing of Isoprene SOA via different pathways may lower the f_{C5H6O+} in the Isoprene-OA factor. Moreover, seasonality may also have an influence on the lower f_{C5H6O+} at the urban sites since the sampling periods at the urban sites are May and August, when the isoprene concentration is relatively lower than that during the sampling periods at the rural sites (i.e., June and July)."

9. In section 4.1.4, the authors discuss the degradation of levoglucosan during atmospheric aging and thus loss of BBOA tracer ion signal. However, aging of primary BBOA emissions has also been shown to increase signal of these tracer ions in the AMS. For example, Heringa et al. (2011) showed that aging of BBOA emissions in chamber studies resulted in the formation of SOA which fragmented to give signal at C2H4O2+. This increase in C2H4O2+ from SOA would offset some of the loss of tracer signal due to levoglucosan oxidation. This point could be also be mentioned when discussing the BBOA factor. (Reference: Heringa, M.F., DeCarlo, P.F., Chirico, R., Tritscher, T., Dommen, J., Weingartner, E., Richter, R., Wehrle, G., Prévôt, A.S.H. and Baltensperger, U.: Investigations of primary and secondary particulate matter of different wood combustion appliances with a high-resolution time-of-flight aerosol mass spectrometer, Atmos. Chem. Phys. 11, 5945-5957, 2011)

Response: Thanks for pointing this out and we have amended the text in the revised manuscript.

"These two ions are largely produced by levoglucosan, which is formed from the breakdown of cellulose in biomass burning (Schneider et al., 2006). In addition, Heringa et al. (2011) showed that SOA produced during the aging of primary biomass burning emissions could contribute to these two ions."

However, even if SOA produced during the aging of primary biomass burning emissions could contribute to these two ions, both laboratory studies and ambient measurements show solid evidence that the signals at these two ions decrease rapidly with photochemical aging. We have amended the text in the revised manuscript.

"It is important to note that the BBOA reported in this study likely only represents the relatively fresh OA from biomass burning. For example, laboratory studies revealed that the oxidation of levoglucosan is fast in both the gas and aqueous phases (Zhao et al., 2014; May et al., 2012; Hennigan et al., 2011). The fast oxidation of levoglucosan can result in the rapid decay of signals at $C_2H_4O_2^+$ (*m/z* 60) and $C_3H_5O_2^+$ (*m/z* 73), causing the mass spectrum of BBOA to lose its characteristic signature. In addition, laboratory studies by Hennigan et al. (2011) and Grieshop et al. (2009) showed that the mass spectrum of OA from biomass burning becomes increasingly similar to that of MO-OOA after photochemical aging. Ambient measurements in the eastern Mediterranean by Bougiatioti et al. (2014) showed evidence that BBOA could be rapidly converted to OOA in less than a day. Thus, aged OA from biomass burning could be apportioned to the MO-OOA factor."

10. From Figure 16S, the NO3,org diurnal has a maximum for CTR_Jun and YRK_July in the mid-morning, rather than in the night or early morning. I would expect NO3,org to peak in the night due to organic nitrate formation from nighttime chemistry. Why is the peak shifted to the mid-morning for these two cases? Also, organic nitrates are a significant aspect of the paper, so I suggest to either move Figure 16S to the main text or include the NO3,org diurnal in the main text, perhaps in Figure 6.

Response: The shift of $NO_{3,org}$ diurnal maximum from early morning to mid-morning is likely due to organic nitrate formation from photooxidation of VOCs in the presence of NO (i.e., RO_2+NO pathway) in addition to VOCs+$NO_3^{\bullet}$ pathway. For the CTR_June dataset, NO concentration increases quickly from ~25ppt to ~275 ppt in early morning (5am to 7am) and decreases quickly back to ~50ppt from 7am to 10am. The relatively high concentration of NO in mid-morning could lead to the formation of organic nitrate when NO reacts with RO_2 radical. This could be supported by that some particle-phase organic nitrate compounds, as measured by a Filter Inlet for Gases and AEROsols Chemical Ionization Mass Spectrometer (FIGAERO-CIMS), show a daily maximum in mid-morning during the SOAS campaign in the SE US (Lee and Thornton, personal communication).

As suggested by the reviewer, we have moved the Fig. S16 to the main text. We also include the following discussion in the revised manuscript.

"Figure 12 shows the diurnal variation of $NO_{3,org}$ based on the NO_x^+ ratio method with an R_{ON} value of 10. For most of the datasets, $NO_{3,org}$ starts increasing after sunset, which is mainly caused by the oxidation of VOCs by nitrate radical at night. The daily maximum of $NO_{3,org}$ appears in mid-morning (i.e., ~8am), which is likely because photooxidation of VOCs in the presence of NO (i.e., RO_2+NO pathway) also contributes to organic nitrate when the NO concentration is highest."

11. The authors state that an FPEAK value of 0 was used in all datasets (P10491), however, in the supplementary information, it is stated that an FPEAK value of 0.2 was used for RS_Jan. This discrepancy should be resolved.

Response: In this study, we performed two types of PMF analysis. One type is PMF analysis on organic mass spectra only (denoted as PMF_{org} in the main text) and the other type is PMF analysis on organic mass spectra together with NO^+ and NO_2^+ ions (denoted as $PMF_{org+NO3}$ in the main text). An FPEAK value of 0 was used in all data sets in PMF_{org}. An FPEAK of value of 0 was also used in all datasets in $PMF_{org+NO3}$, except for RS_Jan, where an FPEAK value of 0.2 was used. As discussed in the SI in detail, an FPEAK value of 0.2 was chosen for RS_Jan after considering the convergence of $PMF_{org+NO3}$ and the correlation between factors from $PMF_{org+NO3}$ and factors from PMF_{org}.

We have modified the sentence for clarity.

"An FPEAK value of 0 is used for all datasets in our PMF analysis on organic mass spectra, ..."

12. TECHNICAL COMMENTS

(1) P10481, L26, P10482, L23, P10483, L28, P10484, L13, P10484, L27, P10486, L2, P10486, L12, P10488, L2-5, P10489, L7, P10489, L10, P10489, L13-14, P10490, L7, P10490, L12, P10490, L13, P10490, L13, P10490, L16, P10490, L16, P10490, L22, P10490, L23, P10494, L5, P10494, L20, P10494, L21, P10497, L4, P10498, L6, P10500, L3, P10502, L10, P10502, L17, P10503, L5, P10504, L13, P10504, L26, P10505, L21, P10505, L22, P10506, L9, P10506, L22, P10509, L20, P10509, L24 and L25, P10510, L21, P10511, L4, P10512, L21-22, P10513, L15, P10513, L19, P10513, L23, P10514, L19, P10515, L6, P10515, L24, P10515, L28, P10516, L1, P10516, L14.

Response: These comments are related to language and we have modified the text accordingly.

(2) Figure 4: Parts (a) and (b) should be same font sizes (e.g., axes).

Response: The changes have been made accordingly.

(3) Figure 5 caption: "Campaign" is misspelled. Figure 11 caption: "Detection" is misspelled.

Response: The changes have been made.

Response to Reviewer 2

1. A main point that should be clarified in the text is the origin and use of boundary layer height (BLH) data. The authors often refer to the BLH diurnal variation as an element that affects the diurnal pattern of the different chemical species observed at the ground. However, very little information is provided on the origin of these BLH data, and on the actual BLH diurnal variation these data show in the region under investigation. More details are definitely needed in the text on this aspect. At least a Figure with the typical BLH diurnal pattern in summer and winter in the region should be included in the supplementary material. This would (possibly) allow the reader to understand the use of the BLH data in generating the results of Figures 3d and 6d. In fact, to my understanding, at present these data are used in an incorrect way. The authors state they multiply the different chemical species loads at the surface (expressed as µg/m3) by the BLH (see e.g. page 10513, line 25), obtaining units of µg/m2. I do not see the physical reason for that. The concept of the BLH dilution effect is that, given a certain load of the chemical component X (expressed in µg), uniformly distributed within a well mixed atmospheric volume V given by [area(m2) x BLH(m)], the concentration of X that is measured at the ground, i.e. X/V, would be higher when BLH is lower (i.e., in the morning/evening), and would be lower when BLH is higher, i.e. at midday. As the authors correctly describe in the text, this effect contributes explaining why, for example, a given component expected to increase in the central part of the day for photochemical processes, can exhibit a flat diurnal cycle (or even be observed to decrease in the central part of the day). So my point is that the authors give the right explanation to justify some of the BLH-driven diurnal patterns observed, but provide the wrong (or insufficiently explained) demonstration for that.

Response: As the reviewer suggested, we have added a figure with the diurnal variation of BLH measured at the CTR_June and added more descriptions of BLH in the revised manuscript.

We agree with the reviewer's interpretation on the effect of BLH on the diurnal variation of a species and it is essentially the same as ours. The interpretation of multiplying the concentration of a compound X (i.e., $\mu g/m^3$) by BLH (i.e., m) is the integrated column concentration of X (i.e., $\mu g/m^2$) from ground to the top of boundary layer over a unit surface area. The value of X*BLH would be conserved if there is no gain or loss of X in the column regardless of the change of BLH. Thus, this value could indicate the net gain or loss of X in the column without the effect of BLH-driven dilution.

We have included the following discussion in the revised manuscript.

"In Centreville, the boundary layer height (BLH) was measured by a ceilometer. The diurnal variation of BLH is shown in Fig. 14. The BLH typically peaks (i.e., 1300m) at 17:00 and exhibits a daily minimum (i.e., 375m) at 07:00. In order to remove the effect of BLH-driven dilution on the diurnal variation of OA, we multiply the OA diurnal profile by BLH. The interpretation of the product of the concentration of OA (i.e., $\mu g\ m^{-3}$) times BLH (i.e., m) is the integrated column concentration of OA (i.e., $\mu g\ m^{-2}$) from ground to the top of boundary layer over a unit surface area, assuming the OA is well-mixed in the boundary layer. The value of OA*BLH would be conserved if there is no gain or loss of OA in the column regardless of the change of BLH. Thus, this value could indicate the net gain or loss of OA in the column without the effect of BLH-driven dilution. As shown in Fig. 3d, the OA*BLH increases rapidly starting at ~7:00 and reaches a daily maximum at ~17:00. The evident peak in the diurnal variation of OA*BLH suggests a substantial OA production in the day, and that the relatively flat OA diurnal variation (i.e., $\mu g\ m^{-3}$) is largely caused by the BLH-driven dilution."

2. The authors often refer to the term 'Brown Carbon' and use 'Brown Carbon' data in the manuscript. As it can be inferred from the text, with this term they refer to the 'Brown-carbon light-absorption' (units m-1). This should be clearly stated in the text and in the relevant Figure showing these data (Figure 7) in order to avoid potential confusion. Additionally, the description of how Brown Carbon Absorption is derived should be improved (Page 10489 lines 11-15). In fact, a) it is not clear how the Black Carbon information from MAAP is combined to the

Aethalometer one to derive the Brown Carbon Absorption, and, b) I doubt that, in the Aethalometer case, 'the measurements under seven wavelengths (i.e., 370, 450, 571, 590, 660, 880, and 950 nm) were averaged to represent the black carbon concentration' as currently stated. Please explain better and give appropriate reference to the methods used to derive the Brown Carbon Absorption data used in the manuscript.

Response: We have replaced "brown carbon" with "brown carbon light absorption" in the revised text and figures. In this study, the brown carbon absorption is measured by a PILS-LWCC-TOC system as described in section 2.3.3. We have clarified the brown carbon measurements in the revised manuscript.

"The PILS-LWCC-TOC system continuously (i.e., 15min resolution) measured the light absorption spectra of water-soluble organic components. The detailed working principle of the PILS-LWCC-TOC system can be found in Hecobian et al. (2010). In brief, water-soluble species are first dissolved in water in a PILS (Weber et al., 2001). The liquid sample from the PILS is then injected into a Liquid Waveguide Capillary Cell, where the absorption spectra are collected over wavelengths of 200 to 800nm. The average light absorption between 360 to 370 nm is used as a measure of brown carbon light absorption."

For the black carbon measurement, please see our response to the comment #4 of reviewer #1.

3. Although the manuscript is generally well written, some parts/sentences could be shortened, improving its readability. Another issue is the quantity and the specific choice of Figures to be included in the main text and in the supplementary material. Some times in the text the authors refer to Figures in the supplementary material as main points of their discussion. This makes the reading not straightforward as it is needed to switch between the main text and the supplementary material several times. To my opinion all the Figures necessary for the main discussion should be in the main text (Figure S14 for example) and the supplement should only provide the material for a deeper investigation/explanation of the results described in the main text.

Response: In the revised manuscript, we have shortened some long sentences and rephrased some sentences to improve readability. As suggested by reviewer #1, we decide to move Fig.

S16 from SI to the main text. We prefer to keep Fig. S14 in the SI since the key point of these plots is the correlation coefficients of species between different sites, which has been shown in Fig. 13.

4. Specific/technical comments

(1) Title. Possibly, given the contents of the manuscript, the title should be modified as 'Aerosol characterization over the southeastern United States using high resolution aerosol mass spectrometry: spatial and seasonal variation of aerosol composition and sources with focus on organic nitrates'

Response: Thanks for the suggestion and we have changed the title as suggested.

(2) Page 10481 Line 6, Page 10490 Line 6, Page 10490 Line 19, Page 10493 Line 1, Page 10496 Line 14, Page 10499 Line 3, Page 10501 Line 29, Page 10502 Line 20, Page 10503 Line 5, Page 10503 Line 8, Page 10505 Line 22, Page 10505 Line 24, Page 10506 Line 3, Page 10506 Line 20, Page 10509 Line 2, Page 10510 Line 2, Page 10510 Line 23, Page 10510 Line 24, Page 10511 Line 14, Page 10512 Line 19, Page 10513 Line 4, Page 10513 line 7-8, Page 10515 Line 28.

Response: These comments are related to language and we have modified the text accordingly.

(3) Page 10481 Line 11: It would be useful to specify in this abstract what 'important' means here, please provide for example data of % contribution of HOA and COA.

Response: We have made the suggested change.

"Hydrocarbon-like OA (HOA) and cooking OA (COA) have important but not dominant contributions to total OA in urban sites (i.e., 21-38% of total OA depending on site and season)."

(4) Page 10488 Line 5: Please, specify the meaning of V mode and W mode

Response: We have made the suggested change.

"We operated the HR-ToF-AMS in two ion optical modes (V and W) with different sensitivity and spectra resolution,…."

(5) Page 10489 Line 5-25: It would be important to know the temporal resolution of the different datasets introduced in this paragraph.

Response: We have added the temporal resolution of all instruments in the revised manuscript.

(6) Page 10494 Line 10-12: It would be useful to know how these averages were obtained (Average of daily averages? Averages of hourly averages? See also my comment above on the temporal resolution of the measurements).

Response: The numbers in Page 10494 Line 10-12 and in table 1 are the campaign-averaged values based on high-temporal resolution data (1 - 60 min depending on instrument). We have added this information in the footnote of table 1 in the revised manuscript.

(7) Page 10494 Line 20-23: BLH also plays a role in the winter-to-summer difference here, not only emissions.

Response: We agree with the reviewer. We have added "or less dispersion". The sentence now reads as "In contrast, at the urban JST site, the NO_x level is elevated in winter compared to summer, indicating more anthropogenic emissions, or less dispersion, in winter at urban sites."

(8) Page 10495 Line 19: Give reference to Fig. 2 here.

Response: We have made the suggested change.

(9) Page 10495 Line 22-23: I'm not convinced entrainment could play a role in this. In fact, it is more likely to act in the first part of the day, when BL grows and intercepts the residual layers aloft (e.g. Curci et al, Atmos. Chem. Phys., 15, 2629-2649, 2015)

Response: Curci et al. (2015) use lidar observations and model simulations to show that the entrainment into the planetary boundary layer (PBL) has significant impacts on the ground-level aerosol concentrations. This supports our hypothesis that entrainment plays a role in the sulfate diurnal variation. We agree with the reviewer that Curci et al. (2015) showed that the entrainment is more likely to act in the first part of the day. However, Curci et al. (2015) studied the effects of entrainment in the Milan urban area, the meteorology and pollutant sources are different from those in the Atlanta area. For example, the Atlanta metropolitan area is surrounded by coal-fired power plants with tall stacks. As suggested by Weber (2003), these tall stacks emit SO_2, which could undergo cloud processing to form sulfate. In the afternoon when the PBL is the highest, sulfate aloft is entrained into the PBL. This hypothesis could be supported by Wagner et al. (2015), who showed evidence of sulfate production above the PBL in the southeastern US in summer time.

(10) Page 10497 Line 15-16: I cannot see this 'clear lunch and dinner feature' here. This is only shown in 3 out of 6 plots and: a) it is not visible in JST_May, b) in RS-Jan the peak is in the morning at 5 a.m. Please explain this point better or rephrase.

Response: We refer the reviewer to the response to comment #7 of reviewer #1.

(11) Page 10498 line 5: Why not referring to Isoprene-OA as IOA as done for all the other categories?

Response: We thank the reviewer for the suggestion. However, there are already a lot of acronyms about OA factors in the literature now. Thus, we prefer to keep the term Isoprene-OA to be more informative.

(12) Page 10498 Line 8: Acronyms should always be introduced first (not all the readers may know IEPOX stands for Isoprene epoxydiols).

Response: We have made the suggested change.

(13) Page 10501 Line 7-14: Could these BBOA differences be also due to the fact that you compare PM1-related data to PM2.5-related ones? Please, comment.

Response: This is one possible explanation and we have included it in the revised manuscript.

(14) Page 10503 Line 11-13: Please, explain better, see also my comment on BLH effect above.

Page 10513 Line 24-26: See my main comment above on this matter. Please give details on how BLH data have been collected, ceilometer instrument used, BLH retrieval. Provide at least a Figure in the supplementary material showing the typical BLH in the region and its seasonality.

Response: We refer the reviewer to the response to your comment #1.

(15) Page 10508 Line 4: Is that from NaNO3 the only possible contamination?

Response: There are other possible contaminations in addition to $NaNO_3$. In this study, we use the concentration of Na as an indicator to show that the potential interference from mineral dust is small. We have modified the sentences for clarity.

"However, due to the transmission efficiency of PM_1 cyclone, PILS-IC measurements might include contributions from particles larger than 1μm (i.e., inorganic NO_3 in mineral dust). Interferences from water-soluble refractory particles (e.g., calcium or sodium nitrate) are likely small given the concentration of sodium measured by the PILS-IC with a PM_1 cyclone, for example, was negligible and mostly below its detection limit (0.07 $\mu g\ m^{-3}$) (Fig. S13)."

(16) Page 10508 Line 27: I just want to highlight here that a variability of 0.11-0.21 means a difference of about 100%!

Response: We thank the review for this note. The large variability is caused by the wide range of R_{ON} values in the estimation.

(17) Page 10509 Line 12: Why do you consider GT_AUG to be in a transition month? Isn't August a summer month (particularly considering that relevant observation started on July 20)?

Response: In this study, all seven datasets are grouped according to the extent of agreement between the NO_x^+ ratio method and PMF method. For summer datasets (CTR_June and YRK_July), the $NO_{3,org}$ estimated from the PMF method falls within the upper and lower bound of the NO_x^+ ratio method. For winter datasets (JST_Nov and YRK_Dec), the PMF method shows good agreement with the NO_x^+ ratio method with an R_{ON} value of 10. For JST_May and GT_Aug, the NO_x^+ ratio method predicts higher $NO_{3,org}$ concentration than the PMF method, which is caused by that PMF method attributes some organic nitrates to the nitrate inorganic aerosol (NIA) factor as discussed in the manuscript. For the convenience of discussion, JST_May and GT_Aug are grouped as "transition month" datasets. To avoid confusion, we have deleted the phrase "the sampling periods of which were between summer and winter".

(18) Page 10511 Line 18-19: Rephrase the sentence to refer to Fig 12 and then address the reader to figure S14 for deeper analysis (if you believe Fig 12 is more 'efficient' than Figure S14 in summarizing the results you are commenting here'….)

Response: We have rephrased the sentence as following.

"Figure 13 shows the correlation coefficients for $NR\text{-}PM_1$ species between ACSM measurements (stationary at the Georgia Tech site) and HR-ToF-AMS measurements (rotating among different sites). Detailed comparisons, in terms of time series and scatter plots, are shown in Fig. S14."

(19) Page 10512 Line 25-27: I cannot see a 'POA' curve in Figure 5

Response: A "POA" line is not included in Fig. 5 because the POA concentration could be easily calculated by subtracting SOA from total OA. In addition, adding a POA curve makes the plot very busy.

5. Figures

(1) Figure 2: it would be useful to also have an additional panel showing the 'absolute' plot (as in Figure 5).

Response: We have included a figure (Fig. 2a in the revised manuscript) to show the concentrations of non-refractory PM_1 species.

(2) Figure 3: Axis Labels and Tick Labels are not readable at all, please increase the character size of all of them. Axis Limits should better be the same in all panels to allow a more direct and straightforward comparison. For Figure 3d see my comments above. I think it can even be removed without losing much of the manuscript significance. Otherwise, carefully describe its content as mentioned.

Response: The axis labels are not readable in the ACPD version. Once the manuscript gets accepted by ACP, we will request that Figs. 3 and 6 take up a whole page and we believe the figures will become readable. We agree with the reviewer that using the same axis limit would allow a more direct comparison. However, the concentrations of species span a wide range for different sites and seasons, so using the same scale would make the diurnal variability less clear. We have addressed your comments regarding figure 3d above (i.e., response to your comment #1).

(3) Figure 5: in the legend Isoprene-OA could conveniently be indicated as IOA for homogeneity. Numbers at the top of panel b are not necessary as also shown in panel a. Possibly you could add error bars on panel a. I cannot see POA line in the Figure.

Response: We prefer to keep Isoprene-OA to be more informative. We have deleted the numbers in panel a as suggested. Adding error bar makes the figure very busy. A "POA" line is not included in Fig. 5 because the POA concentration could be easily calculated by subtracting SOA from total OA.

(4) Figure 6: All the comments for Figure 3 are still valid for this figure.

Response: We refer the reviewer to the response to your comment #5(2).

(5) Figure 7: Please, put labels a) – j) in the plots. Axis Limits should better be the same in all panels to allow a more direct and straightforward comparison (there is no need to reach 70 in the Y axis of panel d, you can leave 'out of scale' values without losing much information. Use 'Brown Carbon Absorption' rather than 'Brown Carbon' in the plot labels (see my comment above).

Response: We have added labels for each sub-panel. We have addressed the comment regarding the axis limit in response to your comment #5(2). In Fig. 7(d), there is actually a split in the left y-axis. We have replaced "brown carbon" with "brown carbon light absorption" throughout the text and figures.

(6) Figure 8: Please, put labels a) – j) in the plots. Axis Limits should better be the same in all panels to allow a more direct and straightforward comparison.

Response: We have addressed similar comments above.

(7) Figure 9: This Figure is not very readable at least in the printed copy I used. Please, try to improve readability/figure resolution. Please, put labels a) – j) in the plots. Axis Limits should better be the same in all panels to allow a more direct and straightforward comparison.

Response: We have addressed similar comments above.

(8) Figure 11: I think the way this Figure is organized is not optimal. You should rather show the AMS-IC value (Y) versus the relevant range coming from the NOX ratio method with the two RON values (e.g. similarly to the vertical lines of Figure 10 but as horizontal lines in the X axis). You can leave info on the two fit lines; correlation R is obviously the same and should not be repeated. The reason of the offset is not clear and should be better explained. Measurement errors are mentioned in the caption but not shown. Please, specify and/or show such errors.

Response: We thank the reviewer for the suggestion. However, we prefer to keep the figure as it is because it already clearly conveys our main points. The 1:1 line is offset by the detection limit of PILS-IC nitrate (i.e., -0.03 μg m^{-3}) for visual clarity. The uncertainty of PILS-IC measurements is about 10% according to Weber et al. (2001). We have added the information in the figure 11 caption.

(9) Figure 12: In the caption it should be 'values are plotted versus the relevant distance of the measurement site from the GT one, where the…'

Response: We have made the suggested change.

Response to Reviewer 3

1. Page 10481, line 14: So far only the AMS and ACSM are mentioned. Therefore, it seems as if brown carbon was measured by one of these instruments. Give the instruments used to measure brown carbon.

Response: The brown carbon light absorption is measured by a PILS-LWCC-TOC system as described in section 2.3.3.

2. Page 10490, line 4: Only AMS publications are listed, but the aerosol community is much bigger. Could the authors list non-AMS publications presenting PMF results (or similar statistical analysis), since it is supposed to be widely applied in the aerosol community?

Response: We have added the following references.

(1) Positive Matrix Factorization (PMF) Analysis of Molecular Marker Measurements to Quantify the Sources of Organic Aerosols. Jaeckels et al. 2007 ES&T.

(2) Receptor modelling of both particle composition and size distribution from a background site in London, UK. Beddows et al. 2015 ACPD.

(3) Advanced source apportionment of size-resolved trace elements at multiple sites in London during winter. Visser et al. 2015 ACPD.

3. Page 10493, line 20: The authors might reconsider the wording. "Nitrate inorganic aerosol particles" would be solely consisting of inorganic compounds including nitrate but especially in the presented case, the aerosol particles consist of inorganic nitrate and organic compounds at *the same time. How about hyphenating to clarify the word associations: Inorganic-nitrate aerosol particles in contrast to organic-nitrate aerosol particles?*

Response: We thank the reviewer for the suggestion. The terminology "nitrate inorganic aerosol (NIA)" is adapted from Sun et al. (2012b). In order to be consistent with literature and avoid confusion, we prefer to keep it as is.

4. Page 10495 line 9: General comment to ALL figures not just Fig. 3: Error bars are missing, which are of specific importance for the interpretation of diurnal profiles. The axes labeling and legend text is rather small. Please try to use the same scaling (or multiplication) for multiple panels in one figure to support easy comparability.

Response: We refer the reviewer to the response to comment #6 of reviewer #1. The figures will become readable in the ACP version.

5. Page 10496, line 6: Many readers might be more familiar with low-volatility (LV-) and semi-volatile (SV-) oxygenated organic aerosol (OOA). Please move the introduction of more-oxidized (MO-) and less-oxidized (LO-) OOA from page 10502 to here.

Response: We have made the suggested change.

6. Page 10498, line 1: This reference might be obsolete. It seems as if the impact of the lower resolution on the number/type of PMF factors was discussed in this publication, but this is not the case. The authors of the cited publication rather recombine two separate OOA factors and proceed with a 2 factor solution (HOA and OOA) for an easier comparison to PMF results of a collocated HR-AMS instrument.

Response: We have deleted the reference as suggested.

7. Page 10498, line 8: What are IEPOX? The authors should give a brief description as well as an explanation of the acronym since not all readers are chemists.

Response: IEPOX stands for Isoprene epoxydiols. We have made the suggested change.

8. Page 10498, line 25ff: Is the difference in fC5H6O+ solely due to rural vs. urban? Could there be an influence from the seasonality in terms of transient (May, August) vs. summer (June/July) months?

Response: We refer the reviewer to the response to comment #8 of reviewer #1.

9. Page 10502, line 26: The ranges given for LO- and MO-OOA are specific to this publication. Taking the cited references into account, it seems to be hard to generate a generality in the range of values. A short discussion on this might be helpful for nonfamiliar readers.

Response: In this study, the O:C ratios are determined by following the latest procedures as recommended by Canagaratna et al. (2015) (i.e., denoted as "Canagaratna method"). Canagaratna et al. (2015) improved the estimation from Aiken et al. (2008) (i.e., denoted as "Aiken method"), which has been widely used in the literature. The Canagaratna method has only been published very recently. Currently, there are only few studies that calculated O:C ratio using Canagaratna method, thus we don't have many literature values to compare with. The O:C ranges of MO-OOA and LO-OOA for multiple ambient measurements in the northern hemisphere calculated using the Aiken method have been reported in Ng et al. (2010) and Jimenez et al. (2009). If we were to calculate the O:C values of the MO-OOA and LO-OOA reported in our study using the Aiken method, our values are consistent with literature values (Ng et al., 2010; Jimenez et al., 2009).

10. Page 10503, line 8: I suspect the authors consider the identification of specific sources of MO-OOA to be challenging but not the identification of the factor itself. In that case, please rephrase the sentence.

Response: The reviewer is correct. We mean the identification of specific sources of MO-OOA to be challenging. We have rephrased the sentence.

11. Page 10505, line 21: As a general comment, please check on your significant figures (digits) throughout the text and in all figures! The text in figure 9 is hard to read! Please reduce to/highlight the most important information (R values). Take the appropriate detection limits into consideration. Include a zero line to guide the eye of the reader. Give the color coding in the figure caption as well, especially, since the legends are hard to read.

Response: We have used two significant digits in figure 9 and increased the text font in the revised manuscript. The correlation coefficients, which are the most important information, are already listed in table 2. Zero line has been added and the figure captions are color coded as suggested.

12. Page 10506, line 4: The authors mention inorganic nitrates other than ammonium nitrate but do not discuss possible compounds and sources. A short discussion maybe taking the results of Alfarra(http://cires1.colorado.edu/jimenez/Papers/Alfarra_PhD%20Thesis_4Chapter4_Labwork .pdf) into account would be very informative.

Response: We thank the reviewer for the suggestion. The Alfarra's thesis shows the NO^+/NO_2^+ of $Mg(NO_3)_2$, $NaNO_3$, and $Ca(NO_3)_2$, which are mainly from mineral dust. However, these species are negligible in PM_1 in the SE US. In addition, the main reason for smaller R_{meas} than R_{AN} is due to variations in instrument performance. Thus, we decide to delete the phrase "contribution from inorganic nitrates other than ammonium nitrate or". The sentence now reads as: "The negative values are a result of smaller R_{meas} than R_{AN}, at times (see Eqn 1), which is likely caused by variations in instrument performance (Farmer et al., 2010; Rollins et al., 2010)."

13. Comment on figures in supplemental material: Please check on readability of figure legends and axes labeling. Please extend the information in the figure captions. Most probably, figures as Fig. S5 are not as self explanatory to a broader community as the short figure caption suggests. Please check on readability of the figures themselves. E.g. the readability of Fig S8 could be improved significantly when multiple panels were used each displaying only a subset of the lines.

Response: We have made the suggested changes for the SI figures. The major changes are listed below.

(1) Extend the information in the figure captions, especially for figure S5.

(2) Increase the font of axis and legends of some figures.

(3) Check the significant digits.

(4) Divide Fig. S8 into multiple panels

14. Technical comments

(1) Page 10483, line 3, Page 10484, line 19, Page 10490 line 7, Page 10498, line 14, Page 10485, line 21, Page 10498, line 20, Page 10499, line 3, Page 10502, line 15, Page 10504, line 12, Page 10504, line 27, Page 10505, line 25, Page 10507, line 11, Page 10509, line 2, Page 10512, line 18, Page 10512, line 21, Page 10519 line 11

Response: These comments are related to language and we have modified the text accordingly.

(2) Page 10498, line 18: Is this a different "R" than the one mentioned on Page 10499, line 15? If not, please move definition of it to first appearance.

Response: We only used Pearson's R in the manuscript. We have made the suggested change.

Additional Corrections

1. We replace the citation
"Guo, H., Xu, L., Bougiatioti, A., Cerully, K. M., Capps, S. L., Hite, J. R., Carlton, A. G., Lee, S. H., Bergin, M. H., Ng, N. L., Nenes, A., and Weber, R. J.: Particle water and pH in the southeastern United States, Atmos. Chem. Phys. Discuss., 14, 27143-27193, 10.5194/acpd-14-27143-2014, 2014."
With
"Guo, H., Xu, L., Bougiatioti, A., Cerully, K. M., Capps, S. L., Hite Jr, J. R., Carlton, A. G., Lee, S. H., Bergin, M. H., Ng, N. L., Nenes, A., and Weber, R. J.: Fine-particle water and pH in the

southeastern United States, Atmos. Chem. Phys., 15, 5211-5228, 10.5194/acp-15-5211-2015, 2015."

Reference

Aiken, A. C., Decarlo, P. F., Kroll, J. H., Worsnop, D. R., Huffman, J. A., Docherty, K. S., Ulbrich, I. M., Mohr, C., Kimmel, J. R., Sueper, D., Sun, Y., Zhang, Q., Trimborn, A., Northway, M., Ziemann, P. J., Canagaratna, M. R., Onasch, T. B., Alfarra, M. R., Prevot, A. S. H., Dommen, J., Duplissy, J., Metzger, A., Baltensperger, U., and Jimenez, J. L.: O/C and OM/OC ratios of primary, secondary, and ambient organic aerosols with high-resolution time-of-flight aerosol mass spectrometry, Environ Sci Technol, 42, 4478-4485, Doi 10.1021/Es703009q, 2008.

Berndt, T., and Boge, O.: Products and mechanism of the gas-phase reaction of NO3 radicals with [small alpha]-pinene, Journal of the Chemical Society, Faraday Transactions, 93, 3021-3027, 10.1039/A702364B, 1997.

Boyd, C. M., Sanchez, J., Xu, L., Eugene, A. J., Nah, T., Tuet, W. Y., Guzman, M. I., and Ng, N. L.: Secondary Organic Aerosol (SOA) formation from the β-pinene + NO3 system: effect of humidity and peroxy radical fate, Atmos. Chem. Phys. Discuss., 15, 2679-2744, 10.5194/acpd-15-2679-2015, 2015.

Bruns, E. A., Perraud, V., Zelenyuk, A., Ezell, M. J., Johnson, S. N., Yu, Y., Imre, D., Finlayson-Pitts, B. J., and Alexander, M. L.: Comparison of FTIR and Particle Mass Spectrometry for the Measurement of Particulate Organic Nitrates, Environ Sci Technol, 44, 1056-1061, 10.1021/es9029864, 2010.

Canagaratna, M. R., Jimenez, J. L., Kroll, J. H., Chen, Q., Kessler, S. H., Massoli, P., Hildebrandt Ruiz, L., Fortner, E., Williams, L. R., Wilson, K. R., Surratt, J. D., Donahue, N. M., Jayne, J. T., and Worsnop, D. R.: Elemental ratio measurements of organic compounds using aerosol mass spectrometry: characterization, improved calibration, and implications, Atmos. Chem. Phys., 15, 253-272, 10.5194/acp-15-253-2015, 2015.

Curci, G., Ferrero, L., Tuccella, P., Barnaba, F., Angelini, F., Bolzacchini, E., Carbone, C., Denier van der Gon, H. A. C., Facchini, M. C., Gobbi, G. P., Kuenen, J. P. P., Landi, T. C., Perrino, C., Perrone, M. G., Sangiorgi, G., and Stocchi, P.: How much is particulate matter near the ground influenced by upper-level processes within and above the PBL? A summertime case study in Milan (Italy) evidences the distinctive role of nitrate, Atmos. Chem. Phys., 15, 2629-2649, 10.5194/acp-15-2629-2015, 2015.

Griffin, R. J., Cocker, D. R., Flagan, R. C., and Seinfeld, J. H.: Organic aerosol formation from the oxidation of biogenic hydrocarbons, J Geophys Res-Atmos, 104, 3555-3567, Doi 10.1029/1998jd100049, 1999.

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, E. J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of Organic Aerosols in the Atmosphere, Science, 326, 1525-1529, DOI 10.1126/science.1180353, 2009.

Ng, N. L., Canagaratna, M. R., Zhang, Q., Jimenez, J. L., Tian, J., Ulbrich, I. M., Kroll, J. H., Docherty, K. S., Chhabra, P. S., Bahreini, R., Murphy, S. M., Seinfeld, J. H., Hildebrandt, L., Donahue, N. M., DeCarlo, P. F., Lanz, V. A., Prevot, A. S. H., Dinar, E., Rudich, Y., and Worsnop, D. R.: Organic aerosol components observed in Northern Hemispheric datasets from Aerosol Mass Spectrometry, Atmos Chem Phys, 10, 4625-4641, DOI 10.5194/acp-10-4625-2010, 2010.

Wagner, N. L., Brock, C. A., Angevine, W. M., Beyersdorf, A., Campuzano-Jost, P., Day, D. A., de Gouw, J. A., Diskin, G. S., Gordon, T. D., Graus, M. G., Huey, G., Jimenez, J. L., Lack, D. A., Liao, J., Liu, X., Markovic, M. Z., Middlebrook, A. M., Mikoviny, T., Peischl, J., Perring, A. E., Richardson, M. S., Ryerson, T. B., Schwarz, J. P., Warneke, C., Welti, A., Wisthaler, A., Ziemba, L. D., and Murphy, D. M.: In situ vertical profiles of aerosol extinction, mass, and composition over the southeast United States during SENEX and SEAC4RS: observations of a modest aerosol enhancement aloft, Atmos. Chem. Phys. Discuss., 15, 3127-3172, 10.5194/acpd-15-3127-2015, 2015.

Weber, R.: Short-Term Temporal Variation in PM2.5 Mass and Chemical Composition during the Atlanta Supersite Experiment, 1999, J Air Waste Manage, 53, 84-91, 10.1080/10473289.2003.10466123, 2003.

Weber, R. J., Orsini, D., Daun, Y., Lee, Y. N., Klotz, P. J., and Brechtel, F.: A particle-into-liquid collector for rapid measurement of aerosol bulk chemical composition, Aerosol Sci Tech, 35, 718-727, Doi 10.1080/02786820152546761, 2001.

Xu, L., Guo, H., Boyd, C. M., Klein, M., Bougiatioti, A., Cerully, K. M., Hite, J. R., Isaacman-VanWertz, G., Kreisberg, N. M., Knote, C., Olson, K., Koss, A., Goldstein, A. H., Hering, S. V., de Gouw, J., Baumann, K., Lee, S.-H., Nenes, A., Weber, R. J., and Ng, N. L.: Effects of anthropogenic emissions on aerosol formation from isoprene and monoterpenes in the southeastern United States, Proceedings of the National Academy of Sciences, 112, 37-42, 10.1073/pnas.1417609112, 2015.