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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol characterization over the southeastern United States using high resolution aerosol mass spectrometry: spatial and seasonal variation of aerosol composition, sources, and organic nitrates" *by* L. Xu et al.

Anonymous Referee #1

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Review of "Aerosol Characterization over the Southeastern United States Using High Resolution Aerosol Mass Spectrometry: Spatial and Seasonal Variation of Aerosol Composition, Sources, and Organic Nitrates" by Xu, L. et al.

GENERAL COMMENTS

The authors characterized submicron non-refractory particles at several rural and ur-

ban sites in the southeastern US during different seasons primarily using aerosol mass spectrometry. Positive matrix factorization was used to apportion the organic aerosol to various sources, which were dependent on location and season. Organic nitrates were quantified using several approaches and determined to be a significant contributor to the total organic aerosol. Overall, the findings of this paper will be of interest to the community and complement the long term measurements conducted in the region. After addressing the comments below, I recommend this paper for publication. The paper is well organized, however, there are many technical corrections to the writing needed and those listed below are not exhaustive.

SPECIFIC COMMENTS

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.

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?

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?

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?

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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. 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?

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?

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

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sites? Could this be an indicator of mixing of the PMF factors in the urban datasets?

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)

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.

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.

TECHNICAL COMMENTS

P10481, L26: Replace “of” with “to the.”

P10482, L23: Replace “has been” with “is.”

P10483, L28: Should it be “. . .in an urban site.”? Suggest to give name of the urban

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site.

P10484, L13: Add “the” before “SEARCH.”

P10484, L27: Replace “of” with “to.”

P10486, L2: Define LT (local time).

P10486, L12: Replace “lines” with “lanes.”

P10488, L2-5: Reword sentence, “Ionization efficiency. . .”

P10489, L7: Change “includes” to “include.”

P10489, L10: Either pluralize “principle” or add “The” to the beginning of the sentence.

P10489, L13-14: Should read, “For the Aethalometer, measurements at seven. . .”

P10490, L7: Should read, “. . .factors with constant mass spectra. . .”

P10490, L12: Remove “and.”

P10490, L13: Sentence should not start with “m/z’s”.

P10490, L13: Should read, “. . .with a signal-to-noise ratio less than 0.2. . .”

P10490, L16: What is meant by, “CO2+ organic ions?”

P10490, L16: Reword sentence starting with, “In addition, for four datasets. . .”

P10490, L22: “Signals” should be singular.

P10490, L23: Remove “that.”

P10494, L5: Insert “the” between “in” and “ith OA factor.”

P10494, L20: Insert “the” between “YRK” and “NOx.”

P10494, L21: Insert “the” between “JST site” and “NOx.”

P10497, L4: Pluralize “emission.”

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P10498, L6: Remove “ion.”

P10500, L3: Insert “the” between “when” and “isoprene.”

P10502, L10: Should read, “. . .based on analysis of ∼900 filters. . .”

P10502, L17: Change “due to that” to “because.”

P10503, L5: Change “atmospheric” to atmosphere.”

P10504, L13: Insert “the” between “with” and “PMF.”

P10504, L26: Monoterpenes should be singular.

P10505, L21: Insert “the” between “using” and NOx+.”

P10505, L22: Should read, “An RON value. . .”

P10506, L9: Insert “The” before “NOx+ ratio method.”

P10506, L22: Change “from” to “in.”

P10509, L20: Add “the” between “from” and “best-estimate.”

P10509, L24 and L25: Change “of” to “to.”

P10510, L21: Should read, “. . .likely compensate for the weaker. . .”

P10511, L4: “disappear” should be “disappears.”

P10512, L21-22: Change to “This likely arises from the fact that biogenic VOCs. . .”

P10513, L15: Change to read, “. . .shows a moderate increase. . .”

P10513, L19: Change to read, “The lack of a prominent daytime increase. . .”

P10513, L23: Add “the” between “of” and “boundary.”

P10514, L19: Insert “for” after “compensates.”

P10515, L6: Insert “the” before “gas-phase.”

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P10515, L24: Remove “of” after “despite.”

P10515, L28: Remove “that” after “by.”

P10516, L1: Should read, “. . .the total OA at the rural YRK site. . .”

P10516, L14: Should read, “. . .resulting in an under-prediction of the pollution levels in urban areas.”

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

Figure 5 caption: “Campaign” is misspelled.

Figure 11 caption: “Detection” is misspelled.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 10479, 2015.