**Atmospheric
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Discussions

Interactive comment on "Seasonal characterization of submicron aerosol chemical composition and organic aerosol sources in the southeastern United States: Atlanta, Georgia and Look Rock, Tennessee" *by* S. H. Budisulistiorini et al.

Anonymous Referee #1

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This manuscript reports year-long measurements of non-refractory submicron particles by Aerodyne Aerosol Chemical Speciation (ACSM) at an urban (JST) and rural (LRK) sit in the southeastern US. Source apportionments of organic material were made for each season at each site. Overall, this paper is well written and addresses a relevant topic within the scope of the journal. The authors should provide details about how the PMF solutions are determined as well as the uncertainty analysis. I disagree with some

of the factor identification and suggest the authors to re-check their analysis carefully. I recommend this manuscript be published after the following comments are addressed.

Specific comments:

(1) Page 22385, line 17-21: Perma Pure PD-50T isn't designed for flows containing particles. Have the authors tested the particle loss through the dryer?

(2) Page 22385 to 22386: The slope obtained from ACSM NR-PM1 + BC and SEMS PM1 is complicated with the CE value. Although a CE value of 0.5 was chosen, substantial differences exist when comparing the ACSM results with other measurements at the sites studied herein (Budisulistiorini et al., 2014). Given the large uncertainty of CE (as well as other uncertainties in the ACSM quantification of mass concentration), I don't think it is meaningful to derive the particle density by this method. Also, the authors estimated the particle density based on average composition. It is perhaps better to use 1.4 instead of 1.2 g cm-3 for organic material based on our current understanding (Hallquist et al., 2009; Kuwata et al., 2012). Nevertheless, to me, 1.48 in Table S3 and 1.75 for summer JST are quite different. The question is what is the reasonable tolerance for density estimates.

(3) Page 22386, last paragraph: The determination of the "best" PMF solution for each case seems quite subjective. Although Figures S2-S9 are provided for the PMF diagnositcs, there is no description about them. It was mentioned that the uncertainties of selected solutions were investigated with the seed, fpeak, and bootstrapping tests. But I can't find any further information. The authors should provide the details about the best-solution determination and the uncertainty analysis.

Some questions regarding the PMF diagnostics are listed below.

– The Q/Qexp values are not near 1, and are different for the JST (2-3) and the LRK (∼0.2) best solutions. Have the errors of the input data and the downweighting treated properly? If the Q/Qexp contributions are dominated by a few temporal spikes or

plumes, test PMF runs excluding those time periods should be performed and analyzed. It is not unlikely that certain plume events represent source profiles very different from the long-term profiles. The authors should make sure those events won't bias the PMF solutions.

– Figure S2: It looks like there are significant Q/Qexp contributions from marker ions of m/z 41 to 60. What does the 5 factor solution look like?

– There are still some patterns in the residual time series for the "best" solutions that need to be checked, for example, March in Fig. S2, April in Fig. S3, earlier October and December in Fig. S5, and mid March in Fig. S6. Those features may indicate that more factors are needed to explain the data. Besides, the season separation may not be ideal and adjustments may be needed.

– The chosen of a particular rotation (e.g., fpeak = -0.15 for LRK spring in Fig. S7 and -0.1 for LRK fall in Fig. S9) needs to be well explained. It looks like the factor mass spectra are highly correlated for those cases. Tuning rotation to obtain more distinct markers like m/z 82 and 91 would bias the PMF analysis.

(4) Page 22388, line 22: It should be clarified in the text or figure caption that some of the data are published in Budisulistiorini et al., 2015.

(5) Page 22389, line 12-14: How big could the under-prediction be if organosulfates are considered? Studies suggest organosulfates accounted for up to 30% of the organic mass and 4-14% of total sulfate (Lin et al., 2012 and references therein). Have the authors done any analysis on the ambient samples at the sites to quantify the amount of organosulfates?

(6) Page 22391, line 22-30: The production of SOA may be balanced out by the loss and hence appeared as day-time valley. It looks like all OOA-factors and inorganic species show similar diurnal patterns that decrease during the day. Could the authors show data like the diurnal PBL height and other meteorological parameters to specify

the main driver of the day-time decrease? Why do the minimum concentrations of nitrate, organic material, sulfate, and ammonium appear at different time (Fig.4a)?

(7) Page 22392, line 19-30: I would argue that the 91Fac reported in Robinson et al., 2011 (Supplementary) is more likely the HOA factor (see the high R2 values in Table S1). The f43/f44 ratio for the Robinson 91Fac factor (1.2) is also much greater than the ratio herein (0.12). The caveat is that f43/f44 is also sensitive (usally) to the rotation. Overall, I tend to agree with the assignment of this factor at LRK as 91Fac, similar to those identified in Budisulistiorini et al., 2015 and Chen et al., 2015. But at JST spring (summer as well), because the small mass fractions of the 91Fac, noisy temporal variations (Fig. 3a), and the high correlation of the mass spectrum of this factor with LV-OOA. I think it is more likely a "split" factor, which needs to be carefully reanalyzed.

(8) Page 22393, line 7-13: The 82 marker seems not present in the IEPOX-OA factor here (Fig. 2b). The spectrum is highly correlated with LV-OOA (R2∼0.9) but less correlated with lab IEPOX SOA (Tables S1-S2). How confident do the authors believe this is not a "split" factor, or something else (e.g., some type of SV-OOA), but a real IEPOX-OA factor? Similarly for the fall, relatively high 82 and m/z 53 (associated with the 82 peak (Budisulistiorini et al., 2013; Chen et al., 2015; Robinson et al., 2011) are missing (Fig. 2d). The authors should carefully check the PMF analysis for the spring and fall.

(9) Page 22395, line 1-6: The 91Fac for LRK fall seems being mixed with HOA although f44 is high. The CxHy+ ion series clearly present, and in particular, m/z 57 stands out. For comparison, biogenic-related 91 factors are expected to have a m/z 53-55 pattern (Chen et al., 2015).

(10) Given the differences in the mass spectra and diurnal profiles of one factor for different seasons (Fig.2), PMF analysis for combined datasets of different seasons should be done to test if the results are robust and meaningful.

Technical remarks: Page 22388, line 26: It is better to specify here as "non-refractory chloride". Page 22393, line 1 and p22399, line 3: "Chen et al., 2014" should be "Chen et al., 2015". Figure 2. The signals for ions above m/z 60 are too low to see. Ion markers (e.g., 82 and 91) are difficult to tell. Either additional figures or amplified right axes are needed.

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