

Interactive comment on “Particle Size Dependence of Biogenic Secondary Organic Aerosol Molecular Composition” by Peijun Tu and Murray V. Johnston

Peijun Tu and Murray V. Johnston

mvj@udel.edu

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We thank the reviewer for excellent comments and suggestions to clarify and improve the manuscript. Reviewer comments and our responses are given below. Line numbers refer to the original manuscript.

Specific comments:

The referee questions the “similarity” of the size distributions for polydisperse samples a and d (Table S1 and Figure S1).

Author response: We recognize the confusion caused by our wording on p. 4 lines 17-19. What we meant to say, and will modify the text accordingly, is that fine tuning

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of the size distribution of the control aerosol was done in order to assure that we had a sufficient aerosol mass concentration at each of the mobility sizes of interest (35, 60, 85, 110 nm) to permit chemical analysis. Fine tuning was required because of the difficulty of generating particles at the smallest mobility diameter using our particular atomizer.

The referee asks about the definitions of entries in Table S3.

Author response: We will modify Table S2 to include the following footnote for the “Unique Molecular Formulas” column saying that these values represent the average and standard deviation from five replicate samples. Each replicate gives slightly different results, which is why it is important to perform several replicates. The difference among replicates arises mostly from formulas that have very low signal intensities. We will also add a new column entitled “Common Molecular Formulas” that gives the number of common formulas observed in all five replicates for a given sample type – these are the formulas that are evaluated and discussed in the results and discussion. This will give the reader a better idea of repeatability of the experiment, and will show that most of the assigned formulas are indeed detected in all five replicates.

Referee comment concerning page 7 lines 4-11 and Figures 2a and 2b.

Author response: We will modify the legend and caption of the figures as well as the text to more clearly indicate the identities of the markers (NAMS, HRMS(+) and HRMS(-)), for both size-selected and polydisperse aerosols. (By the way, the referee got the representations correct in their comment.)

Referee comment about page 9, lines 10-12, why is it “not surprising” that there is no composition dependence for negative ions in Fig. 6b?

Author response: We will expand this discussion in the revised manuscript to explain more thoroughly, while at the same time removing the “not surprising” phrase. Our group and others (e.g. Hall 2013, Tu 2016, Mutzel 2015) have noted in the

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past that molecular formulas obtained from negative ion spectra are generally more highly oxygenated/oxidized than those obtained from positive ion spectra. Highly oxygenated/oxidized formulas are suggestive of molecules that have very low volatilities. If essentially all of the monomers detected in negative ion spectra are nonvolatile, there will be no particle size dependence in their relative ability to be incorporated into particles – all of these molecules will condense with similar probability when striking the particle surface. In contrast, the next paragraph (p. 9 lines 13-22) discusses positive ion monomers, which potentially have a very wide range of volatiles – some are non-volatile, while others are semivolatile. The relative amounts of non- vs. semi-volatile monomers will change as a function of particle size for the reasons given in this paragraph.

Referee comment about Figure s2.

Author response: The caption to Figure S2 will be modified – this spectrum is for 60 nm size-selected particles.

Referee comment about Figure S3. Author response: Labels displaying the specific mass loading values will be added.

Typographical changes noted by referee: Author response: All will be corrected as mentioned by the reviewer. Thank you very much for pointing these out.

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