

Interactive comment on “Size and composition measurements of background aerosol and newparticle growth in a Finnish forest during QUEST 2 using an Aerodyne Aerosol Mass Spectrometer” by J. D. Allan et al.

Anonymous Referee #1

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This manuscript reports an experimental study of new particle growth in a Finnish forest using an AMS. The number distributions, chemically-resolved aerosol mass distributions and mass spectra of ambient particles during event and non-event days are studied and compared. Evidences are shown that organic species played important roles in new particle growth. Compounds that are possibly involved are proposed. These results are novel and provide valuable insights into the growth of new particles in forested atmospheres. This paper is also well written and I recommend publication

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after the authors address following comments.

General comments:

I think the AMS wire data and related discussions are not directly relevant to the main findings/points presented in this paper. While it is important to report the use of different collection efficiencies (CE) in this study, the wire data shown in Fig. 1 and the discussions made in Page 8763 do not directly support the use of specific values (i.e., CE = 0.5 for dry particles & CE = 1 for wet particles). In addition, particle bounce on the vaporizer, which is mentioned as a favored hypothesis in page 8763, does not seem to be consistent with the phenomenon shown in Fig 1. Within this context, I think it will be helpful that the authors explain and discuss Fig 1 in more detail. Or if it is difficult to find a convincing and consistent explanation I'd like to suggest that Fig 1 is included in supplementary information.

A main focus of the paper is to investigate the roles played by organic compounds in new particle growth and the compounds that were possibly involved. The authors certainly have done a very good job discussing the major findings and hypothesis. However, I am very curious to see an explicit comparison between the mass spectra of organic aerosol from event days to those from non-event days. (Figure 4 only shows the speciated mass spectra of continental influenced air mass, and only m/z 25 - 90 is displayed.) I was also wondering about a couple of details regarding the AMS organic data; 1) how many organic fragments were scanned for size distributions during this study? and 2) what are the average size distributions of these m/z 's during event and non-event days? This information needs to be presented either in the main text and/or as supplementary information.

It also puzzles me why standard EI mass spectrum, rather than AMS spectrum, of verbenone is presented, especially since the two spectra are likely to be very different? In terms of presentation, it will be helpful that minor tick markers (better of 1 unit m/z increment) are shown in the mass spectra(Figs. 4, 8, 10).

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In addition, I think it is worthwhile to demonstrate the correlation between the DMPS and AMS size distributions in Figs 6 and 7 by 1) including the DMPS particle volume (or mass) image plots and 2) scaling the y axes of the image plots by assumed particle density so that D_{va} is related to D_m .

Specific comments:

Page 8758, line 4, remove “the” in front of “each”.

Page 8758, line 25, change “they” into “the”.

Page 8759, 2nd sentence, to my knowledge, Zhang et al. (ES&T, 2004) reported that the composition of the growing particles was predominantly ammonium sulfate at the beginning of new particle growth in Pittsburgh, but oxygenated organic species was a major component of the new particles several hours later, due to condensational growth.

Page 8759, line 24, remove “has” in front of “was”.

Page 8761, 3rd sentence of the 3rd paragraph, I don’t think that “the AMS data provide the direct linkage between data from the sizing instrument and the chemical information from offline analyses”. Based on my understanding, the AMS itself provides information on the size resolved composition of non-refractory species in particles. One has to assume density, particle shape and the contributions from refractory species to relate data from an AMS to that from the sizing instruments (e.g., DMPS). While it is possible to relate some offline analysis results to sizing data (for example based on mass closure calculation), it is hard to believe that a “direct linkage” can be provided. Please clarify.

Page 8761, line 29, please consider revising “Particles less than the lower limit”.

Page 8764, last sentence of 1st paragraph: please specify the RH ranges corresponding to collection efficiencies of 0.5 and 1, respectively.

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Page 8766, line 7, insert Dva in front of $<200\text{nm}$.

Page 8766, a side-by-side mass spectra comparison of organics during non-event vs. event days will be helpful.

Page 8768, line 8-10, I'd like to point out that the oxygenated organic signal at m/z 30 (CH_2O^+) may not necessarily be a small feature in mass spectra of ambient or lab organic aerosols.

Page 8769, lines 8 and 9, I have trouble understanding "the instrument response is largely invariant at alkane $\text{C} \# > 10$ ". Is it according to the appearance of the mass spectra?

Page 8770, lines 10- 12, please clarify "the relatively abundances of carbon, hydrogen, and oxygen are conserved in the fragments". I also have trouble understanding why "the low m/z peaks could be indicative of terpene oxidation products"? Please clarify.

Page 8770, 2nd paragraph, another possible reason for the lack of organic mass in particles $< 200\text{nm}$ at low concentrations of monoterpene oxidation products could be related to the size distribution of available surface area for condensable species. It will be helpful that the authors clarify this point by examining the condensational sink of accumulation mode particles in comparison to that of particles $< 200\text{nm}$ during periods when concentrations of monoterpene oxidation products were low.

Page 8772, please revise the first sentence of the 2nd paragraph. To my knowledge, the organic AMS signature during event days presented in this study has been observed in several rural/remote locations. These results have been presented at conferences although I am not aware of the publication of similar results.

Page 8780, Fig 1, please define the x axis. What are the "sectors" corresponding to? I'd like to suggest the use of more viable units (such the ratio of the distance from the edge/center of the vaporizer to the diameter of the vaporizer), rather than arbitrary units.

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Page 8784, I think it will be helpful to show the side-by-side average AMS size distributions during event and non-event days as well.

Pages 8785-8786, Figs 6 & 7, why are the images so pixilated? If any averaging is applied to the AMS data, please mention it in the text and/or in the captions.

Page 8786, please define the parameter that is plotted in Fig. 7a, is it total organic mass or total mass?

Page 8787, caption of Fig. 8, the use of “sources” here is rather vague.

Page 8788, 2nd sentence of the caption of Fig. 9, check subject and verb agreement.

Page 8789, fig. 10, while it is true that the high temperature evaporation process of organic compounds in the AMS may cause larger extent of fragmentation and low relative intensity of larger fragments compared to NIST MS, the degree of deference is very much dependent on the structure of the compounds. Large m/z fragments of oxygenated compounds with aromatic ring can be prominent in the AMS mass spectra. So, in order to support the major statements relevant to Fig. 10, it will be helpful to show the AMS mass spectrum of verbenone if possible.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 8755, 2005.

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