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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.

J. Smith (Referee)

jimsmith@ucar.edu

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General Comments:

The following are comments on the paper "Size and composition measurements of background aerosol and new particle growth in a Finnish forest during QUEST 2 using an Aerodyne Aerosol Mass Spectrometer" by Allan et al. This study attempts to identify the species responsible for particle growth following new particle formation in a forested

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environment, and is a valuable contribution to the literature on the subject. Among the most important observations made (in the opinion of this reviewer) are:

1. There is a unique spectral fingerprint of organic aerosol seen in Hyytiälä, and this seems to be the same in the larger accumulation mode (condensed onto background aerosol) as with the Aitken mode (condensed onto particles form by nucleation).

2. Particles that grew following nucleation (sub-100 nm diameter) seemed to be dominated by organics.

3. There is maybe some evidence of organic nitrates present in particles, in addition to aliphatic compounds and OVOCs that may suggest oxidized monoterpenes.

Specific Comments and Technical Corrections:

Specific questions and comments are presented below. While I am mostly satisfied with the main conclusions of the paper, I am somewhat worried about the parts of the paper that address the "and new particle growth" part of the title.

8758, Line 4: remove "the" before "each"

Line 4-5: "and can be of either" does not fit (grammatically) with the earlier clause of the sentence. Consider revising to "which can be" or "and these can be".

Line 18: "size" is ambiguous here. Consider revising to "mobility diameter".

Line 20: Consider revising "compositional nature" to simply "composition".

Line 25: A comment here on the lower size limit and required mass for ultrafine particle (low pressure) impactors would seem appropriate here. Also there have been some recent advances in the area of offline analysis in which investigators use "aerosol concentrators to collect and analyze particles as small as 10 nm aerodynamic diameter with collection times of 3 hours and more. See, for example, the papers by Geller et al.

8759, Line 5: Please confirm where a binary mechanism is actually proposed in the

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Kulmala 2004a reference. I don't believe any statement is made regarding the mechanism of nucleation other than the belief that it is decoupled from growth.

Line 7: check tense agreement of "occurs".

Line 22: Consider revising "different to urban" to "differ from that in"

8759: The top paragraph on this page, which is an introduction to the site and the study, seems out of place from the other two paragraphs which provide background information on similar studies. Consider changing the order such that the top paragraph is the last one in the section, placing the other two background paragraphs more appropriately with the background material on offline (impactor-based) techniques.

8760, Line 23: consider changing "aerosol particles" to simply "particles".

8761, Line 5: Consider deleting the word "derived".

Line 9: For this sentence (which continues a point made by the sentence that precedes it), it is not clear how molecular decomposition on the vaporizer is an exception to the bias towards low molecular weight peaks caused by the temperature of the vaporizer. If I understand this correctly these two sentences seem to be making the same point. If so consider combining into one.

Line 13: is "ensemble" necessary when this is inherent in the meaning of the word "aerosol"?

Line 18: Regarding the point made on the power of size-resolved composition in this sentence and the one that follows the authors state a "direct linkage" and "assignment" of particle sizes determined by the AMS to those obtained through other means, but as I understand it the vacuum diameter derived from the AMS requires knowledge of particle density in order to make correspondences with the mobility or aerodynamic diameter. Please discuss in the text (or just clarify my misunderstanding!).

Line 28: Here I assume "size" refers to "diameter"?

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8762, Line 6: I suggest changing the first of 3 “it was” appearances in this paragraph with “the instrument” or something similar. The use of passive voice is especially apparent in this paragraph.

8763, last paragraph: Fig. 1 seems to lend strong support to the notion that the particle beam indeed diverges significantly at low RH. This paragraph suggests that this is not in agreement with the modeling studies from the Univ. of CO or light scattering studies performed at Aerodyne/Boston Col. The authors seem a little too quick to suggest that beam divergence is not a factor at all, however Fig. 1 still seems to me to support that view (would not particle bounce result in just an overall reduction in number, still preserving the high-RH distribution?). If particle bounce is the hypothesis favored by the authors, please comment on the significance of their observations in Fig. 1.

Line 10: please comment on the size range of the particles plotted here. Is this all ambient aerosol? If so, then perhaps the results plotted here are weighted by number concentration and may not be generally applied to a study that focuses on the chemical properties of ultrafine aerosol.

8764, Line 19: consider changing “aerosol particles” to simply “particles”. Line 23: Here I believe you are referring to the mass concentrations and size of the background aerosol. Please clarify.

Figure 2: As in the text that refers to the figure, I am not sure if “arctic outbreak” means just the background aerosol during the days in which the wind blew from the north, or if “outbreak” refers to new particle formation. Please state this clearly. It is interesting to me that sulfate is so dominant in the particles from these northern air masses (and that the particles are so acidic). Is this observation supported by other studies?

Line 27: Similarly, I am not sure if the text and Fig. 3 refer to the composition of the background aerosol (i.e., the particles that existed prior to, and during, nucleation).

8765, Line 5: Forgive me for not having a good meteorological background, because

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(as in my comment above) when I hear the word “outbreak” I think about the sudden increase in particle number concentrations that characterize a new particle formation event. In this case I think the authors are again referring to air masses that originate from Europe. Please consider changing the terminology.

Line 15: I don’t understand this sentence. Is there a clause “Ěwhen the wind is blowing from the continent.” missing at the end of this sentence?

Line 25: this sentence of course begs the question: did back trajectories or other meteorological measurements support the idea that the emissions were relatively constant during nucleation and growth?

8766, Line 13: In this paragraph, please identify the prevailing wind direction for each of the days plotted in Figs. 6 and 7.

Figure 6: As in the preceding comment, please note the prevailing wind direction in the caption if you think it’s important. It’s not clear from the figure that the composition of the sub-100 nm particles during 18:00 - 00:00 GMT (Fig. 6c) is appreciably different from the composition of the same class of particles during 00:00 - 6:00. I can certainly appreciate that a lack of particulate mass can make the chemical characterization difficult, but don’t the plots here mainly support the observation that the background aerosol can collect organics during the course of several hours? If I am reading plot (a) correctly I see that there is again not a clear correlation between organics concentration and the DMPS Aitken mode. Please comment.

Figure 7: The chemical composition data suggest that this is an example of a northern air mass. Are the contours the same as those in the legends of Fig 6? Please state this in caption. Again there is not a clear correlation between the organic aerosol mass and the growth mode from the DMPS data, which is somewhat disappointing.

A thought about Figs. 6-7: Perhaps it makes more sense to plot the DMPS data in terms of dM/dlogDp (i.e., mass distribution, making some assumption of bulk particle

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density). If this is done then it may address my disappointment in not seeing a nice correlation between the AMS chemical data and the physical DMPS data.

Line 21: It's not clear to me what particle size range corresponds to the spectrum plotted in Fig. 8. Is this an average of all particles sampled during the growth event? Of so, can the DMPS data be used to tell us how much of the data shown here can be related to the accumulation mode as compared to the Aitken mode?

Line 25: Just a point for clarification: the aliphatic sequence was observed in the smallest particles as well as the larger ones? By stating that the fingerprint was invariant, does that also mean that the relative abundances of the aliphatic peaks to the unsaturated and/or oxygenated peaks are invariant with particle size? Over what size ranges was this invariance observed?

8768, Line 16: I very much enjoyed reading the insights of the authors presented in this section. Whereas I am not sure that the data show, unequivocally, that the particles that were involved in growth were organic (it seemed to me that the low signal-to-noise ratios may add a degree of uncertainty to this particular finding), I will not argue on semantics in the opening sentence of this section (but would nonetheless request that the authors comment on the role of uncertainty in their findings!).

Line 20: I would argue that perhaps the observation that the chemical signature is the same between the accumulation mode and the Aitken mode may in part depend on the size ranges considered for each. Certainly, in talking about the growth of very small particles (sub-20 nm), one cannot imagine that more volatile compounds (e.g., nitric acid and lighter OVOCs) would participate in growth. On the other hand, if you consider that the AMS chemical signatures are weighted by mass towards the larger diameters in the Aitken mode then perhaps it is not too unreasonable that the signatures are similar. It would seem important to qualify the findings of this study with the size range of the particles measured, if known.

8769, Line 9: When it's stated that the instrument's response is invariant as a function

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of carbon number, this would imply that the signal from a C20 is half that of a signal from a C10. Is this correct? If this is so, then why can't the AMS still obtain information on the carbon number? Is the signal too weak?

8771, Line 4: The point made here regarding the AMS response to monocarboxylic acids brings to mind an important issue regarding the AMS that might be suitable for discussion in this forum. To my knowledge there are no published studies in which the response of the AMS has been calibrated with laboratory-generated particles of known composition (and here I'm especially thinking about organics). I realize that there must be studies like these performed, and hopefully they have been peer-reviewed and easily available to the community. Can you please refer me to such studies? Better still, can you briefly summarize the expected response of the AMS to the compounds of relevance to the current study? For example, is it known whether alkanes are more easily ionized and detected as compared to ketones or aldehydes? What is known about the detection limits of carboxylic acids?

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