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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The role of VOC oxidation products in continental new particleformation" *by* A. Laaksonen et al.

A. Laaksonen et al.

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We thank referee #2 for the useful comments.

General comment: The manuscript is well written, but it summarizes too a large degree previously published results. This is the weakest point of the manuscript. I suggest that the reviewing part should be strongly reduced, it is clear from the previously published evidence that at least in later stage of particle growth OVOC contribute significantally. The added value by the new data and observations should then be stronger highlited, e.g. the assessibility of 10nm particles by the e-Tandem DMA measurements. I suggest publication after the major changes proposed above. In any case the authors should consider the major comments and correct the minor points.

Answer: We disagree with the referee somewhat. To summarize the logic of the anal-

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ysis of particle chemistry, we 1) infer detailed information of the chemical nature of condensed organics from the various large particle analyses, 2) show that the AMS mass spectra are consistent with the large particle results and that the mass spectra in the largest (below one micron) and in the smallest (below 100 nm) size ranges detected by the instrument are similar, and 3) use the OTDMA to show that the ethanol growth factors of 50 nm and 10 nm particles are both correlated with MTOP during nucleation events and therefore it is highly likely that they both contain similar organics which can also be found in larger particles. While most of the results of 1) and 2) have been published earlier in the QUEST special issue, we believe it is the integral approach of considering all the results together that is important here. We therefore would prefer retaining (the rather compact) descriptions of the previously published results. We agree with the referee, however, that the new observations could be highlighted more, and will try to improve the ms accordingly.

Major comments and questions p. 7825, line 17f The formulation that "the growth factor depends on the relative size difference of the molecules in the particle and ethanol" is misleading: do you refer to the "usual" that for a given dry volume, the "number of soluble entities" depends on the density and the molecular mass of the solutes or do you refer to the finding of Petters et al. 2006 (Tellus, 58, 196-205) that dissolution of macromolecules leads to specific deviations from ideality? The latter could be indeed understood a effect of relative size of solute and solvent.

Answer: This is just bad formulation. What we intended to say was simply that if we have two particles having the same dry size, one composed of small molecules and the other of large molecules, both taking up ethanol so that the resulting ethanol mole fractions are the same, then the particle containing small molecules shows a larger GF. We will reformulate.

p. 7826, line 20 ff This section is difficult to understand. What is a more southerly sector. What is a continental outbreak, how is it related to the southerly sector. Is there a northerly sector, is that related to arctic outbreaks? Please specify.

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Answer: Continental outbreak means air masses arriving from the European continent, i.e. southerly direction. There are Arctic outbreaks from the northerly direction. Will be clarified.

p. 7827, line 20 ff Doesn't the appearance of unusual organic signatures in AMS indicate that growth is not driven by MTOP and sesquiterpene oxidation products ?

Answer: As discussed in Allan et al (2006), based on searching in the NIST mass spectral data base, the pattern of observed m/z intensities matched that of a specific terpene oxidation product (e.g. verbenone - that was identified in off-line analysis), directly relating the AMS organic signature to terpene oxidation products. That signature is unusual only because the Allan et al (2006) results from Hyytiala were among the first AMS observations in a remote (boreal) forest environment.

p. 7828, line 18, Table 1 Were such high particulate concentrations of pinonaldehyde commensurable with the pinonaldehyde mixing ratios in the gas phase? Or was the particulate phase supersaturated with respect to the gas phase?

Answer: See our reply to Neil Donahue.

p. 7829 line 22, and 7831 line 1 Were the altitude distributions of VOC and SO2 measured during the flights? Is there more experimental evidence than the height profile of the particle concentration for the contribution of VOC oxidation products to particle growth? Every ground near source could explain the simple fact of height dependence.

Answer: Unfortunately, neither SO2 nor VOC height profiles were measured. However, we would like to point out there are no strong SO2 sources near the operation area of the aircraft, at least ones that could cause SO2 concentrations to decrease uniformly with height. Furthermore, sulphuric acid concentrations in Hyytiälä are generally so low that they can only explain a minor fraction of observed particle growth (e.g. Boy et al., ACP 5, 863, 2005).

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Minor points: p. 7825, line 2 normal AMS do not use thermal desorption, but flash evaporization. Which technique was applied ?

Answer: "Thermal desorption" will be replaced with "fast thermal desorption (600C oven temperature)"; Thermal desorption and flash vaporization are equivalent; the former is "fast" at high temperature. We agree it is important to report the oven temperature.

p. 7824, line 1 ff Sellegri et al. 2005a, and 2005b is quoted in the text, but only one Sellegri et al. 2005 appears in the reference list.

Answer: Will be fixed.

p. 7827 line 13 misplaced "(13,14)"; ?

Answer: Will be fixed.

Figure1 and 2 Is the deviation of the intercepts from 1 significant ? Does it mean anything ? Errors margins are missing.

Answer: We do not believe that the deviation is significant, there is too much scatter in the data to make such an inference. The main point is that these correlations only appear during nucleation events. We will add error margins.

Figure 5 What is shown in Figure 5, number concentrations of all particles or number concentrations between 3-10 nm?

Answer: All particles, this will be clarified.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 7819, 2007.