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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Overview of the field measurement campaign in Hyytiälä, August 2001 in the framework of the EU project OSOA" *by* M. Boy et al.

Anonymous Referee #2

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General comments:

This manuscript serves as an overview of the filed measurements made during the Hyytiälä campaign, one of the two field campaigns as part of the EU project OSOA. Comprehensive instrumentation was employed to measure the closely coupled physical, chemical, meteorological systems of interest; the novel Organic Tandem Differential Mobility Analyzer was particularly impressive and yielded important results. Different aspects of the measurements were discussed in detail, including observed aerosol nucleation and growth, aerosol flux and vertical profiles, measurements of ozone, SO2, NOx and other precursors, chemical characterization of photo-oxidation products of VOCs in the gas-phase and aerosol-phase, as well as meteorology and solar radia-

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tion. The data analyses, overall, are of good quality, and many results are helpful to further our understanding of the very complicated sources and formation pathways of secondary organic aerosols. However, some data interpretations are problematic and some statements are misleading, which requires clarification or re-phrasing. The use of the English language is at times awkward, which may have partly caused the problems mentioned above. I will next provide my specific comments, followed with suggestions on some technical corrections.

Specific comments:

1>. In section 2.1, paragraph 2, the authors state that Śthe DMPS system used here actually consists of two systemsŠ. It would be to the readersŠ benefit if the authors can describe how these two systems are positioned in the sampling line, i.e., is one upstream of the other, and if so, which one is at the upstream and why?

2.> In section 2.8, the authors state that Śa backup filter was placed behind the front filter to determine possible sampling artifacts (correct the spelling!)Š. However, no further discussion of sampling artifact appears later in the manuscript. It would serve the purpose of ŚoverviewŠ better if the authors can discuss this in some more detail, or at least point the readers to relevant papers.

3>. In section 3.2.1, paragraph 2, the authors state that Śthis result (from Gao et al., 2001) seems to be in contradiction to our measurementsŠ and suggest Śother parameters such as the number concentration of pre-existing particles may be more important for the formation of new aerosols than the variation in SO2 concentrationŠ. After examining Gao et al., 2001, I find this argument somewhat problematic, mainly because the particle nucleation and growth scenario there is very different from the one studied here (the real atmosphere), which essentially renders these two observations not directly comparable.

In Gao et al., the pre-existing particle concentrations were probably almost constant for the four cases (SO2 <0.1, 0.5, 2.5, and 6 ppb), since the chamber was routinely

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cleaned before each experiment and the initial particle concentration was always below 0.1 m-3. And even if some products of α-pinene-ozone may have been involved in particle nucleation, possibly serving as pre-existing aerosols, they were still probably quite constant for the four cases judged from the initial gas concentrations. The relatively constant T and RH almost certainly cannot explain the observed difference in the particle nucleation behaviors in the four cases. However, during the Hyytiälä campaign, parameters such as T, RH, solar irradiance, pre-existing particle concentration, and precursor gas concentration may all have varied substantially from day to day in the lower atmosphere. In such a non-controlled environment, as the authors themselves stated elsewhere in the manuscript, more than one parameter may have played concerted roles in particle formation. No single parameter can be concluded or precluded as the sole, or major, contributor to the observed aerosol nucleation, at least not based on the evidence presented here. In summary, I do not believe the observations in this paper and in that of Gao et al. are directly comparable. The authors should clarify on this.

In addition, the SO2 concentration was below the DL of 1.34 ppb (the 1st sentence in paragraph 2 needs clarification, by the way) during the Hyytiälä campaign. Have the authors considered the likelihood that sub-ppb level of H2SO4, being oxidized from SO2, and especially in the presence of ammonia, could have potentially explained the nucleation they observed [Korhonen et al., 1999]?

4>. A related concern occurs in the 2nd paragraph in Śsummary and conclusionsŠ. The authors conclude that Śconcentrations of all carboxylic acids would be high enough to fulfill the role of condensing species on the thermodynamically stable clusters during the time of particle burstsŠ. An important question left unanswered here is Ű what are these TSCs during the Hyytiälä campaign and, how are they formed initially? Since the authors propose in the end that Śproduction of newly formed particles is homogeneous in the PBLŠ, the origin of TSCs becomes a crucial issue, which the authors may want to address to some extent. Interestingly, the ubiquitous presence

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of SO2 and NH3 in the lower troposphere makes H2SO4, from oxidation of SO2, a likely source for TSCs, later growing into larger aerosols after condensation of organic vapors. After obtaining Śvertical profiles of aerosols throughout the day with size and number distributions and simultaneous measurements of different gas speciesŠ, as suggested by the authors, it may be worthwhile, among other things, to look at the effect of SO2 on nucleation by studying cases with other parameters being comparable (constant).

5>. In section 3.3.1, paragraph 1, it is stated that the ŚECPLUCŠs results indicate a predominance of formic acid in the forested area of HyytiäläŠ, whereas Śthe UVAR measured concentrations of formic acid mostly lower than acetic acidŠ. This is, however, a significant discrepancy, since the different measurement results would suggest different dominant sources. The authors should address why this discrepancy occurred, and discuss which one more likely represents the real values in the field campaign.

Suggestions on technical corrections:

1>.Abstract, paragraph 1, suggest changing to ŚAs part of the OSOA projectŠ.

2>. Abstract, paragraph 2, lines 19-20, suggest changing to Śhigher concentrations OF pinonicĚ.in the gas phase, which indicates A preference to the particle phaseŠ.

3>. Abstract, paragraph 2, line 23, change to ŚĚreached values around 19% of the sampled aerosolsĚŠ.

4>. Abstract, paragraph 3, line 5, suggest changing to Śthe results give first indications that production of new aerosols happens throughout the PBLĚŠ.

5>. Section 1, paragraph 1, line 3, change to ŚĚsecondary organic aerosols BY applying a combination of ĚŠ; line 8, change to ŚĚ a better scientific understanding of the sourceĚŠ.

6>. Section 2.3, paragraph 1, line 9, change to Śtemperature and pressure measure-

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mentsŠ.

7>. Section 3.1.1, paragraph 1, line 2, change to Śin August and in less clear patternsĚŠ; paragraph 2, last line, change to ŚĚthe very small newly-formed particlesŠ.

8>. Section 3.1.3, paragraph 5, line 8, change to Śunlike the hygroscopicityĚŠ.

9>. Section 3.2.1, paragraph 2, line 2, change to Śbelow detection limit of 1.34ppbĚŠ; line 12, change to Śnewly-formed particlesĚŠ; line 15, change to ŚĚother parameters such as the number concentrationĚŠ; last line, change to ŚĚthe variation in SO2 concentrationŠ.

10>. Section 3.3, paragraph 1, suggest changing to ŚĚthe gas-phase and particle-phase components were performedĚŠ; perhaps change toŠĚstudy the spatial distributionĚŠ.

11>. Section 3.3.1, paragraph 1, line 17, change to Śthat favours acetic acidŠ; line 19, change to ŚĚburning (Talbot et al., 1987). This seems not to have happened in theĚŠ.

12>. Section 3.3.2, paragraph 1, line 2, change to Śmonoterpene-skeletonedŠ; line 15-16, change to ŚFor pinonic, nor pinonic and pinic acids, concentrations in the particle phase were higher than the correspondingĚŠ; line 21, change to ŚĚthe atmospheric concentrationS of OH radicals and ozone HAVE an effect onĚŠ.

13>. Section 3.4.1, paragraph 2, line 4, suggest changing to Śassuming that the ratioĚŠ.; paragraph 4, lines 3-4, suggest changing to Śon average, the total concentration of PAH in the Finnish samples was lowerĚ from the previous short-termed experimentĚŠ; paragraph 7, line 8, change to ŚĚhas been suggested to ĚŠ.

14>. Section 3.4.2, title, change to ŚDetermination of photo-oxidation products in PM2 and PM2.5Š; paragraph 4, last line, suggest changing to Ś2 time that of pinic acidŠ; paragraph 5, line 7, change to ŚĚcontains a carboxyl and a per-ester functionĚŠ.

15>. Section 4, paragraph 1, last sentence, change to Śthese results point more to

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the conclusion thatĚŠ; paragraph 2, line 2, changes to ŚĚthe sum of terpenes FROM the number concentrationĚŠ; last paragraph, line 6, changes to ŚĚprofiles of monoterpene concentrations showedĚŠ; line 11, changes to ŚĚdifferent parameters such as temperature, humidity, ĚŠ; last line, changes to ŚĚof concentrations of different gas speciesŠ.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3769, 2003.