Atmospheric Chemistry and Physics Discussions

Interactive comment on "Applying the CPCB setup to study the hygroscopicity and composition of freshly-formed 2–9 nm particles in boreal forest" *by* I. Riipinen et al.

W. Birmili (Referee)

birmili@tropos.de

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General recommendation

The authors provide a new study on the properties of fresh nucleation mode particles. Elucidating the mechanism(s) how these new particle are formed in the atmosphere has occupied a lot of researchers for many years, and a definite single answer how the process works seems still not within sight. Therefore this article covers an important subject.

Multiple measurement techniques were deployed at the well-established Hyytiälä site in the Finnish boreal forest. The CPC battery, introduced in an earlier paper, is applied

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to a more extensive data set, and combined with another quite new instrument, the NAIS. The scientific data evaluation and interpretation is sound, although I am asking for several clarifications and improvements noted below. The text is very well written and I enjoyed reading it. I see this article as an important step towards a further understanding of the particle formation process in the boreal forest region.

As a whole I recommend publication in ACP once the below issues have been addressed.

Specific comments

1) Aerosol inlet: When measuring particles as small as 2 nm great care is needed for an accurate and artifact-free measurement. It is quite easy to introduce unwanted nanoparticle losses (diffusion, electrostatic precipitation), for instance, when using excessive lengths of tubing, or introducing turbulence in manifolds. You can even have significant losses towards building walls if the inlet is not far away from that building. In many of the papers on Hyytiälä I am missing a good description of the inlet systems used. This also includes, for instance, the height at which the measurements were made (below or above the canopy, how far from the ground?). Also, I assume that NAIS, DMPS and CPCB used different inlets. A description would be really useful to assess the quality and comparability of these nanoparticle measurements. As an improvement I suggest to add a small chapter 2.1.4 that summarizes some of the above mentioned key properties of the aerosol inlets.

2) Although this information might be contained in some of the references, I am missing an explicit statement on the reference instrument regarding the CPC efficiency calibration. Probably an electrometer? So please indicate which type, model, and sampling flow.

3) The CPC efficiency curves seem to change with time, although not to an extreme extent (Figs. 2 and 3). Changes are probably related to different environmental conditions, but due also to short-term and long-term instabilities in the temperature control

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circuits of the CPCs, parts of the CPCs like the wicks and saturators undergoing an aging process... Maybe we are not even capable of better defining the CPC efficiency curves than you showed. A conclusion, however, is that we are only able to define the CPC efficiency with some uncertainty. As we can see from the Figs. 2 and 3, the uncertainty does not relate to a simple parameter like the cut-off size or the overall amplitude, but even the shape of the curve seems to be variable.

An implication is that the data processing procedure (i.e. determining ambient cut-off diameters, such as described in section 2.2.) is inflicted with an inherent accidental error. Whether this error is relevant or not could be elucidated by a sensitivity study. What I am asking you is to try to estimate the propagation of error in the CPC efficiency curve into the D50 that is determined later for the ambient aerosols. You could use the experimental CPC efficiency curves, and add some uncertainty that changes the curves' shape according to the differences that occurred between the two calibrations. The resulting error in D50 should be stated as an inherent uncertainty of the method, and taken into account during the discussion of the results.

A similar sensitivity study would be desirable in relation to the DMPS data. I am aware that the DMPS data were probably corrected for all possible losses inside the instrument and inside the aerosol inlet (please confirm, whether this is true). Nevertheless, there might still be a fundamental error attached when determining the size distribution. As an example, the charging efficiency of 3 nm particles is known only at an absolute accuracy of $\pm 50\%$. Therefore, the real values of 3 nm particle concentrations measured by the DMPS might be higher or lower by that amount. It would be useful to know how such an error would propagate into the D50 determined as well.

The sensitivity studies described would generate more confidence in the accuracy of D50. The resulting uncertainties should be noted in the conclusions, and maybe included in some of the Figures as well.

4) Were D50 cut-off sizes determined for completely soluble or organic particles?

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5) NAIS: You state that the NAIS cannot detect ambient particles smaller than 2 nm due to interference with its own charger ions. Using a NAIS, we have made the same practical experience ourselves. In Kulmala et al. (2007a), Figure 1, one can see in NAIS measurements a steady population of particles below 2 nm, which is interpreted as stable molecular clusters. In your Figure 7 we see a steady population of particles at the lower end as well — Unfortunately I cannot see at which diameter because of the missing axis ticks and labels. To me, the findings/statements seem contradictory. Please clarify which observations in the NAIS plots probably correspond to charger ions, and which not.

6) The "Results and Discussions" section would be much more readable if it were divided into subsections using appropriate headers.

7) Please expand the conclusions section. Currently, this includes only very scarce "conclusions". Here one would like to learn more details on how well you can actually discriminate totally soluble particles from organic particles, ambient particles (mixture of all), and insoluble particles.

Formal issues:

Title: The term "CPCB" should be spelled out, because it is not an everyday term. The word "setup" is superfluous. "Hygroscopicity and composition": 1) Hygroscopicity measurements are commonly associated with a humidification of particles below supersaturation. Therefore the term "hygroscopicity" may be misleading. An alternative may be "activation property", or "water affinity". 2) How confident are you that you are really looking at (chemical) composition? Looking at chemical composition would require different substances, compounds or classes of compounds to leave fingerprints in the data, which could be uniquely attributed to specific compounds. From a proper study of "chemical composition" I would also expect the presentation and/or discussion of corresponding compound calibrations of the instrumentation, which seems to be not the case. I therefore recommend to rethink the title in view of all these points.

p. 14895, l. 23: use simple past instead of present perfect.

Same page, l. 25: Use the definite article like "The proposed atmospheric..."

p. 14896, l. 7. Maybe the authors can include another relevant citation on the subject (HAFEX), where statistically relevant annual cycles of newly formed particles, their growth rates, and calculated product rates of VOCs are shown (http://www.atmos-chem-phys.net/3/361/2003/).

p. 14897, l. 29: Insert a reference (no citation) to the TSI Inc., with the location of their headquarters.

p. 14898, l. 12: Delete the citation Kulmala and Hari (2005) here since it gives the misleading impression that the CPCB data was collected during those authors' work.

p. 14898, l. 20: "Material" (uncountable noun) should be singular.

p. 14898, l. 23: Replace "were made... match" by "were matched".

p. 14901, l. 14: The NAIS "was" or "has been" developed.

p. 14902, l. 12. Misleading sentence. I guess you mean something like "Because of minor drifts... concentrations ratios were normalised to a ratio of unity, based on night-time measurement values when the smallest particles ($< 10\,\mathrm{nm}$) were overwhelmingly absent."

p. 14909, l. 9: Clarify what you mean by "role of organics": Mass fraction of the particles, or maybe mass fraction of the condensing material?

Figure 1: The sketch is irritating because some lines signify real aerosol sampling flows, while others imply rather associations with labels. Try to keep the real aerosol flows as lines but disconnect the other labels. "Aerosol in" could be replaced by "Ambient aerosol", "x nm" by something like "$D_c = $ x nm".

Figs. 2 and 3 might be combined as Fig. 2a and b.

Figure 4 could be made much clearer. Also add tick labels for particle diameter axes.

Figure 5: Avoid axis labels that span multiple graphs.

All Figures: Try to use always the same labels, for instance either "TSI 3785", or "CPC 3785" etc. Currently the labels differ quite a lot.

Figure 5: Labels are misleading, use, for example, "DMPS (simulating CPC 3010)".

Figure 7: Ticks for the diameter axis are sadly missing. Also add tick labels at the bottom and the top of that axis.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 14893, 2008.