Atmos. Chem. Phys. Discuss., 9, S2205–S2211, 2009
www.atmos-chem-phys-discuss.net/9/S2205/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Charged and total particle formation and growth rates during EUCAARI 2007 campaign in Hyytiälä" *by* H. E. Manninen et al.

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Received and published: 20 May 2009

Answers to the referee comments by Anonymous Referee #1 on our manuscript "Charged and total particle formation and growth rates during EUCAARI 2007 campaign in Hyytiälä" by H. E. Manninen et al.

We thank the referee for the constructive comments to help us to improve the manuscript. Our detailed answers to the comments are as follows.

Specific comments 1. 2nd paragraph of the introduction should be swapped with the 1st one. Would be great to reference few studies on the growing list of locations where NPF was observed. Regarding Amazonian forest it is not clear whether measurements were not performed there yet or they were, but NPF was not observed.

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We modified the revised manuscript as suggested. We added references to three new locations where NPF have been observed (Beijing, Wu et al., 2007; Southern African Savannah, Laakso et al. 2008; Himalyas, Vendaz et al., 2008). We also clarified the statement about observing NPF events in Amazonian forest. The sentence in question was modified to "The Amazon rain forest seems to be the only place so far where new particle formation (NPF) and growth has not been observed when measured with proper instrumentation."

2. Instrumental section should clarify better, why CPCB is able to measure particles down to 2nm when compared to DMPS system. There is also confusion with the statement that CPC cut-off diameter was 3nm. I believe it is due to the effect of using water CPC in CPCB. Otherwise, CPC used with butanol should be very similar in CPCB and DMPS systems. This issue is very central in later analysis. For instance, is it possible that systematic biases between CPCB and DMPS could artificially produce particles in 2-3nm range? Was there any specific control of the instruments to avoid such a possibility? The range of DMPS system is more often assumed to be up to 800nm as few uppermost channels are not reliable due to noise problems (poor statistics).

We clarified section by adding some sentences: "The CPCB consists of four Condensation Particle Counters (CPCs, see e.g. McMurry, 2000): one butanol- and water-CPC pair with cut-off diameter 3 nm calibrated for insoluble silver particles (TSI-3776, TSI-3786 [UWCPC]) and the other pair with cut-off diameter 6 nm for silver particles (TSI-3772, TSI-3785). The idea behind CPCB is to study the water solubility of nanoparticles, having the same calibrated cut-off diameters for water- and butanol-CPCs in the case of insoluble particles. According to laboratory calibrations, the CPCB is able to detect aerosol particles as small as 2 nm in diameter (Kulmala et al., 2007b) when the solubility of the nucleation mode particles to the condensing vapor of the CPC results in a decrease of the heterogeneous activation diameter inside the CPC. Water-CPCs detect hygroscopic particles down to lower particle sizes compared to butanol-CPCs due to increased activation probability in water vapor. Respectively, the activation probabil-

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ity of lipophilic particles is higher in butanol vapor than in water. During the NPF events in Hyytiälä, the smallest particles activate for growth at smaller sizes in water than in butanol vapor (Riipinen et al., 2008). Before and during measuring in the field, the cut-off size of each CPC was adjusted by varying the temperature difference between the saturator and the growth tube with laboratory generated insoluble silver particles (Petäjä et al., 2006)."

We admit that measurements of sub 3 nm atmospheric particles are challenging. Therefore, we used two different methods to determine their concentration and evolution namely NAIS and CPCB to avoid artifacts; also both instruments were calibrated under laboratory conditions. We calculated the number concentration of 2-3 nm particles as a concentration difference between the CPCB and DMPS readings, when the UWCPC cut-off size during NPF event is assumed to be close to 2 nm (Riipinen et al., 2008), whereas the DMPS detection limit is 3 nm. We also calculated the number concentration of 2-3 nm particles from number size distribution measured by the NAIS. The results for formation rates at 2-3 nm agreed well based on both measurement methods. We clarified the revised manuscript accordingly.

3. Author should provide more information on classification of event days into subclasses rather than stating them as Ia, Ib and II. What does it exactly mean "applicability e.g. to a growth rate analysis"?

We clarified classification of event days into subclasses as follows. "The event days were classified further into subclasses Ia, Ib and II according to the shape of the growing nucleation mode and possibility to use the size distribution data in further study of the event day, for instance, in growth rate analysis. Class Ia events had a clear continuous shape as the formation of particles and subsequent evolution towards large sizes continued for several hours. In class Ib events the clear and continuous growth did not start at cluster sizes or the growth was suppressed. Class II events had unclear shape of growing mode indicating inhomogeneity of the studied air mass." The term "applicability e.g. to a growth rate analysis" refers to the possibility to use size distribution data

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in further study of the event day, for instance, in growth rate calculations.

4. Please give reason why NAIS data was not used in growth rate calculations, when all other instruments were.

The actual growth rates were analyzed using AIS, BSMA and DMPS data. In principle NAIS can also utilized for this. The preliminary analysis using NAIS data gives corresponding values for growth rates. However, since NAIS is relatively new instrument to measure number size distribution of neutral and charged atmospheric clusters and particles, we have not yet performed full calculations for growth rates based on NAIS data in total particle mode. We have calculated charged particle growth rates based on AIS data which corresponds to NAIS data measured in ion mode. The corona charger ions in NAIS still limit the total particle growth rate calculations in the size range 1.3-3 nm. We added following sentences to revised manuscript: "In principle the NAIS can also be utilized for growth rate estimations. The preliminary analysis using NAIS data gives corresponding values for growth rates."

5. Why the effect of coagulation was neglected in the growth rate analysis and what were those conditions allowing to do so?

We assume that this comment refers to p. 5127, l. 5-6. In this study the coagulation is included in the analysis because we are calculating growth rates from the measured data i.e. from observations. Atmospheric observations include all aerodynamic processes (both condensation and coagulation) growing the nucleation mode. Therefore, we reformulated the paragraph as follows to clarity our wording. "The growth rates estimated in this study are directly calculated from the observed number size distributions i.e. apparent growth rates (e.g. McMurry et al., 2005; Stolzenburg et al., 2005). The apparent growth rate includes condensational growth due to various vapors as well as growth due to both internal and external coagulation. Condensation and self-coagulation represent real growth of the particles, whereas external coagulation shifts the nucleation mode towards larger sizes by favoring the smallest particles in the mode

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and apparently growing the geometric mean diameter of the mode. This process is strongly dependent on the coagulation sink provided by the pre-existing population. However, coagulation is very likely to be insignificant under the conditions encountered during the study period (Kerminen et al., 2004)."

6. Why GR3 in the size range of 1.3-3nm was calculated from BSMA data and not AIS as they covered the same size range?

Actually, the growth rates of the charged particles was estimated in all three classes - 1.3-3 nm, 3-7 nm and 7-20 nm - from the AIS data and in the two first classes from the BSMA measurements. We assume that this comment refers to p. 5128, l. 5-6 where the method for calculating total and charged formation rates was described. We decided to use GR3 values calculated from the BSMA data because the BSMA is considered to be more sensitive when measuring in cluster ion size range. As the growth rate dominates formation rate calculations, we wanted to use the same GR when we calculated formation rates for different measurement methods. Thus we can say that difference in formation rates calculated from different measurement data is not just due to different GR values for different instruments.

7. Sensitivity analysis showed that estimation of coagulation sink had the highest impact on growth rate uncertainty. The assumption of coagulation error of 10% therefore needs strong justification.

The estimated error by a factor of 2 in growth rate is mainly due to observational uncertainty, and the factor of 2 is certainly maximum value for that. Coagulation sink is not the one, which has main impact. The error in the coagulation sink of 10% is mainly due to experimental error related to sink determination and DMPS measurements. We clarified the origin of the uncertainties to the revised manuscript.

8. In the case of ion-induced formation rates, the median value of 10% should be accompanied by the uncertainty range of 1.7. That would put some of the other studies in better agreement with this study which should be discussed as well.

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We included following uncertainty estimations to the revised manuscript: "The median value for daily fractions of ion mediated nucleation is 10 % during spring 2007 in Hyytiälä. As uncertainty estimation for contribution of ion mediated nucleation to NPF, we took into account the uncertainty range of 1.7 for ion-induced formation rates and 1.9 for total formation rates. Incorporating the estimated uncertainty ranges into the total and charged particle formation rates, we estimate that, as a median, the ions and their recombination products can explain about 9-15% of the particle formation. Furthermore, the recombination rate used in the calculations was the maximum, which results in an overestimation of the ion-contribution. In addition, there are several days when ion contribution is dominating (2 day during this period). "

Technical corrections: p.5122. 10-15 lines. Style should be improved by removing repetition of "on the other hand".

We agreed, and modified the revised manuscript as suggested. We simply removed the phrase "on the other hand" from line 10 to avoid repetition.

p.5131 line 19. replace "indicating" with "which suggests".

We modified the revised manuscript as suggested.

p.5132 line 11. replace "different approaches" with "different measurement approaches" as it relates to different instruments I believe.

We assume that this comment refers to "different methods". We modified the revised manuscript accordingly. We also replaced "different methods" from p. 5131 l. 21 with "different measurement methods" to uniform terminology.

Added references to the revised manuscript

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