

## ***Interactive comment on “ Seasonal variation of CCN concentrations and aerosol activation properties in boreal forest” by S.-L. Sihto et al.***

**S.-L. Sihto et al.**

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The authors gratefully acknowledge the comments by the anonymous referee 1.

We give detailed point-by-point answers in the text below. The referee comments are presented in boldface and the corresponding replies as normal text below each comment.

### **Anonymous Referee 1**

**The authors present measurements of cloud condensation nuclei (CCN) in a boreal forest, near Hyytiälä, Finland. They determine the activation size threshold**

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**of the particle by two ways, i.e. by determining the fraction of particles that activate and based on measurements of the hygroscopic growth factor at a relative humidity (RH) = 90%. They find reasonably good agreement between these two methods. The extensive data set, acquired over a year of measurements, allows investigating the seasonality of CCN activation parameters (fraction, critical diameter, hygroscopicity) but results show quite consistent average monthly values. However, significant differences are seen during new particle formation events. The main results of the study are the facts (i) that – on average – CCN hygroscopicity can be described by  $\kappa = 0.18$  which is in agreement with previously determined  $\kappa$  for mixed SOA/inorganic particles, (ii) new particle formation events change the fraction of activated particles due to the formation of many small particles and (iii) growth of newly formed particles into CCN sizes occurs over the span of a day. These results are useful and should be highlighted more clearly throughout the manuscript. I recommend publication after consideration of my additional comments below.**

**General comments - The analysis of both CCN and growth factor data relies on the fact that all particles are internally mixed. Are there composition and mixing state measurements in similar locations available that support this assumption? The width of the measured growth factor distribution might give a hint of the mixing state of particles. Can this be quantified?**

All reviewers have correctly pointed out that the internal/external mixing was not adequately discussed in the paper. This has been amended in the revised version.

The HTDMA is one of the best instruments to measure the extent of internal/external mixture of an aerosol population. However, the application of the entire growth factor (GF) distribution to the dry size distribution is much more complex than using only the average GF as we have. Kammermann et al. (2010) did a sensitivity analysis on different ways to calculate CCN number from HTDMA and SMPS data, and found that ignoring particle mixing state did not affect CCN predictions. Thus, we decided to use

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only the simplified model. However, the limitations should have been discussed in more detail, and this has now been corrected.

Hyytiälä is a relatively remote site, with no large particle sources close by. The HT-DMA data show more externally mixed aerosol in winter, and is most strongly visible in the range where the Aitken and accumulation modes overlap, i.e. at 75 and 110 nm dry sizes in this study. In summer, the aerosol is less externally mixed, likely due to constant biogenic SOA condensation onto all particles.

**The fact that the same critical diameter is derived based on growth factors and CCN measurements suggests that the hygroscopicity ('kappa') is constant over the RH range of 90% to supersaturated conditions. Usually, kappa derived from CCN measurements are higher than those derived at lower RH since either organics dissolve if more water is available and/or inorganic compounds further dissociate (e.g., Petters and Kreidenweis, 2007).**

We agree with the reviewer, and this issue has already been mentioned in section 4.2.2. In summertime the aerosol in Hyytiälä is more organic-dominated, but on a yearly average the inorganic concentrations comprise a third to half of the aerosol mass. The amount of additional organics dissolving above 90% RH may simply not be high enough to be visible in this comparison.

**Is anything known of the composition (hygroscopicity) of newly formed particles? Can an ageing time scale' be given of these particles until they exhibit the same hygroscopic properties as larger, older particles?**

Ehn et al., 2007, studied the change in hygroscopicity during the growth of freshly formed particles from 10 to 50 nm in more detail. Typically new particles reach 50 nm during the following night. As the same organics that grow the 50nm particles are also condensing on larger particles, it is believed that the new particles are very similar to the older particles by the next morning after formation.

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**Specific comments p.28232, l. 5: Define 'DMA' p.28232, l. 5: replace 'cloud droplet activation' by CCN activation' as 'cloud droplets' implies that a dynamic system exists and not equilibrium conditions.**

Done.

**p. 28233, l. 29: Define 'aerosol activation quantities'**

They are now explicitly given in the text.

**p. 28234, l. 4: add year of experiments (and remove in Section 4.1.) p. 28236, l. 7-10: What are the size ranges and size cuts for 'large' and small' particles?**

Added.

**p. 28237, l. 12: Are there studies that show that surface tension of organics can be neglected? If so, they should be cited here. p. 28237, l. 16: The Köhler equation also includes an osmotic factor (phi) that takes into account the activity of the solutes. The product of phi and number of ions for ammonium sulfate is usually smaller than 3 (Low, 1969).**

This section was rewritten, which renders these comments redundant.

**p. 28240, l. 24: Add reference for the analysis of different air masses.**

There are no published composition data over a whole year to directly show the discussed changes, but as written, "we expect to have lower contribution of SOA" in winter, we do not feel that a reference is needed as the biological activity naturally is higher in summer than in winter.

**p. 28244, l. 26: kappa = 0.6 for ammonium sulfate is certainly at the upper end of determined values (as it assumed full dissociation). How would the results shift if smaller values (e.g., 0.53 (Petters and Kreidenweis, 2007)) are applied?**

This only leads to a change from 84% to 81%. More discussion on the uncertainties

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related to the calculations was added, and this change is small in that respect.

**p. 28245, l. 9: The density of organics is usually around  $1.4 \text{ g cm}^{-3}$ . Thus, the conversion from mass into volume fractions would even yield smaller mass fractions.**

Organic matter in ambient aerosol particles has, on average, lower density than corresponding inorganic matter. As a result, organic mass fraction is generally lower than the corresponding volume fraction. This was added the text.

**p. 28245, l. 29: based on Figure 9, that the critical diameters of  $S = 0.4\%$  and  $S = 0.6\%$  seem nicely to encompass the value of 70 nm, and not only being 'broadly consistent'. It might be useful to give average values here for the critical diameters at these two  $S$ .**

The authors feel that the average can be obtained from the figure with enough accuracy. No actions taken.

**p. 28253, l. 8: Both Figure 2 and Figure 3 do not show a 'significant' change in critical diameter. The day-to-day variability (Fig. 3) seems much more significant than the variation in the averaged values.**

It is Fig. 4 that we refer to with the inter-annual changes in  $D_{\text{crit}}$ , and we feel that they are large enough to be called significant and should be taken into account in parameterizations etc. Naturally the day-to-day changes are also important, but are considerably harder to take into account in any model description of CCN activity.

**Figure 5: Increase symbol size and connect legend to caption (what is meant by 'min', 'max', 'mean'?)**

The caption was unclear, and has been rewritten concerning the min/max values.

**Figure 6: Add, a, b, c, d, to the figures and use in text.**

Done

C15006

**Technical comments p. 28233, l. 29/30: change word order: 'how are CCN concentrations affected: : '**

Done.

**p. 28235, l. 21: DMA has been defined before**

Second definition removed.

**p. 28235, l. 24: Define 'DMPS' (and remove definition a few lines later)**

Rewritten.

**p. 28235, l. 26: Introduce 'RH' here**

RH is now defined already on line 20.

**p. 28236, l. 3: add 'a' (in a size range: : :) p. 28240, l. 3: Replace 'kappa' by Greek letter. p. 28240, l. 19: add 'a' (: : : to be a larger difference: : :) p. 28246, l. 5: Add 'the' (in the atmosphere) p. 28247, l. 9: replace 'inside' by 'during'**

Done or rewritten.

**p. 28248, l. 18: replace '??A1' by 'A1'**

This was actually supposed to be Fig. 7 and has been corrected.

**p. 28248, l. 24: Use NPF instead of 'new particle formation' p. 28250, l. 17: replace 'less big' by 'smaller' p. 28252, l. 26: 'campaign' Table 1: Footnote is redundant Figure 1, caption: Replace 'with' by 'at' (: : :at five water supersaturations)**

Done.

References

Ehn, M., Petäjä, T., Aufmhoff, H., Aalto, P.P., Hämeri, K., Arnold, F., Laaksonen, A. and

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