Atmospheric
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Discussions

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Interactive comment on "The analysis of size-segregated cloud condensation nuclei counter (CCNC) data and its implications for aerosol-cloud interactions" *by* M. Paramonov et al.

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Anonymous Referee #1

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The authors present a collection of long-term CCN measurements sampled at the SMEAR II station in Finland with the outlined goals of investigating temporal variations of aerosol properties and CCN behavior, determining the aerosol mixing state and its seasonality, comparing this study's long-term measurements with shorter-term measurements made in several other published studies, and providing comprehensive

insight into aerosol-cloud interactions in the boreal environment.

This paper is well written and presents an impressive 29 month dataset of CCN properties. Though it doesn't present significant advances in its methods of data collection and analysis, the comprehensive CCN dataset presents a fine overview and comparison of CCN-relevant aerosol properties in the boreal environment. The paper is well cited and its aims are clearly defined, though some specific items should be further addressed.

Response: The authors of the current manuscript would like to sincerely thank the referee for the constructive comments, criticism and suggestions. All of the comments have been carefully considered and addressed, and responses can be found below after each comment.

Major revisions

1) The title and aim (iv) indicate that a comprehensive insight of aerosol-cloud interactions and its implications will be addressed. At its current state, I would consider it at most a qualitative discussion on potential aerosol-cloud interactions. A discussion of the importance of the paper's findings and the impacts of ignoring said findings on droplet number would be interesting and show the potential importance of capturing CCN behavior in the boreal forest environment. Without these elements or something similar, I would not consider implications on aerosol-cloud interactions to be one of the key points in this paper.

Response: The authors of the current manuscript do agree that the broader concept of aerosol-cloud interactions is not properly addressed in the paper. While the discussion of aerosol-cloud interactions in a boreal environment would most certainly improve the quality of the paper and properly correspond to the title, such discussion, unfortunately, is not possible in its entirety. This primarily stems from the fact that no measurements of cloud microphysics were carried out in the analysis and no data on cloud droplet number concentrations were available; the paper concentrates on CCN only. In order

to avoid misguiding the reader, the title has been changed to reflect the implications of analysed CCNC data for the cloud droplet activation, instead of aerosol-cloud interactions. This notion has also been corrected in the title of the manuscript and elsewhere throughout the paper.

2) Seasonal and monthly median κ values as high as approximately 0.8 are shown with error bars extending up to 1 (e.g., Figures 6 and 8). This is an extremely high κ for an area typically dominated by organics, and the reason for this is not discussed in the paper. An analysis of chemical composition is necessary to provide a clear analysis (Figure 8).

Response: The referees and the reader need to remember that the median κ of 0.74 in February is for particles measured at Seff of 0.1% only – those with diameters $\sim$150 nm. This value of 0.74 excludes the hygroscopicity of all smaller particles, which were measured at higher levels of Seff. If the size/Seff is disregarded, the median κ in February across all Seff levels becomes 0.3. The confusion related to this value is another reason why the first author of this manuscript believes that the use of κ derived from CCNC data may be misleading and needs to be used carefully, always indicating which Seff levels were used in the instrument setup. This goes back to one of the main conclusions about the variation of κ distributions with size and the necessity to always indicate κ as a function of Seff. Some more discussion has been added to the corresponding section, and the paragraph in page 9702 has been altered to read: "It is plausible that the more active SOA formation and the increased organic fraction being the result of increased emissions of the volatile organic compounds (VOCs) from the surrounding boreal environment in the summer are responsible for reducing aerosol hygroscopicity when compared to the winter time. Considering that in Fig. 8 the Dc and κ points for Seff of 0.1% mirror each other, it is important to point out that the aerosol hygroscopicity can be inferred from the critical diameters alone. The median κ of 0.74 in the month of February for Seff of 0.1% is very likely related to a higher mass fraction of sulphate within the aerosol mass and a generally slower growth of

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particles to larger sizes, which allows for a longer time for the oxidation and aging of particles. What is more important to remember is that this value of κ only reflects the hygroscopicity of larger particles, those with diameters of $\sim$150 nm. Naturally, as the size decreases, the hygroscopicity decreases (Table 1), and the seasonal pattern disappears. The winter peak in aerosol hygroscopicity presented here agrees well with seasonal patterns presented by Pringle et al. (2010) and Sihto et al. (2011) for sites in Germany and Hyytiälä, respectively..."

Minor and technical revisions

1) Page 9688, line 14 – Which T is used (e.g., mid-column temp or average temp)?

Response: Average T. The sentence now includes: "T is the CCNC column average temperature recorded for each spectrum"

2) Page 9691, line 18 – I recommend clarifying that 1 μm is the cutoff at which a particle is considered a CCN.

Response: It is not very clear what is meant by this comment. In the text 1 μm refers to the size of droplets after the particles are exposed to a certain supersaturation. The growth to such sizes allows for easier detection and counting by the OPC. The sentence has been modified to read "The typical residence time of the aerosol particles in the saturator unit is on the order of 10 sec – sufficient enough for the resulting drops to grow to $\sim$1 μm in diameter."

3) Page 9691, line 29 – Does "full scan" mean the full scan of size-segregated CN by stepping through all of the sizes? It should be clearly stated to avoid confusion since the previous two sentences discuss non-size-segregated measurements.

Response: The authors believe that the sentence "The full scan at one Seff takes, on average, 17 minutes 40 seconds, with activation spectrum taking 17 minutes." makes it clear that the discussion is about both size-segregated measurements (activation spectrum) and non size-segregated measurements (additional 40 seconds). No changes

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have been made.

4) Page 9692, line 14 – You state that dataset has undergone rigorous procedures in order to remove bad data. Please expand upon the procedures used (e.g., counting statistics, unstable supersaturations) in addition to what is discussed in Section 4.

Response: The following sentence was added to the corresponding paragraph: "The dataset was scanned for unstable Seff levels, problems with CPC and OPC and the resulting counting errors of NCN, NCCN and A, and differences between set and measured sheath flow rate."

5) Page 9699, line 19 – If you are referring to the same average value of 0.18 on line 18, "median" needs to be changed to "average." It also makes sense to report this work's average when comparing with other published averages (e.g., page 9697, line 25).

Response: The sentence was corrected to indicate that the hygroscopicity parameter kappa of 0.18 as reported by Sihto et al. (2011) is indeed an average and not a median. The overall median kappa of 0.22 in this study was also inserted into the corresponding sentence.

6) Page 9703, line 17 - Cerully et al. (2010) should be Cerully et al. (2011).

Response: Corrected.

7) Page 9727, Figure 7 - While the figure provides a nice picture of the spread in the dataset, I would suggest using the data to quantitatively calculate the chemical dispersion in terms of $\sigma(\kappa)$ and $\sigma(\kappa)/\kappa$ as was done in Cerully et al. (2011) and Su et al. (2010).

Response: The referee suggestion to include the chemical dispersion $\sigma(\kappa)$ to demonstrate the degree of heterogeneity in the chemistry of particles would certainly increase the knowledge of the variability of the measured κ values in a quantitative manner. However, besides warranting the inclusion of the description, derivation and applicabil-

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ity of $\sigma(\kappa)$ in the manuscript (which would increase the length of the manuscript), the authors wonder whether such analysis would substantially improve the conclusions about the variability of κ beyond what is already said and depicted in Figure 7. For the purpose of this paper, the qualitative depiction of the spread in the distributions of κ values, as done in Figure 7, is deemed sufficient.

References Cerully et al.: Aerosol hygroscopicity and CCN activation kinetics in a boreal forest environment during the 2007 EUCAARI campaign, Atmos. Chem. Phys., 11, 12369–12386, doi:10.5194/acp-11-12369-2011, 2011. Su et al.: Hygroscopicity distribution concept for measurement data analysis and modeling of aerosol particle mixing state with regard to hygroscopic growth and CCN activation, Atmos. Chem. Phys., 10, 7489–7503, doi:10.5194/acp-10-7489-2010, 2010.

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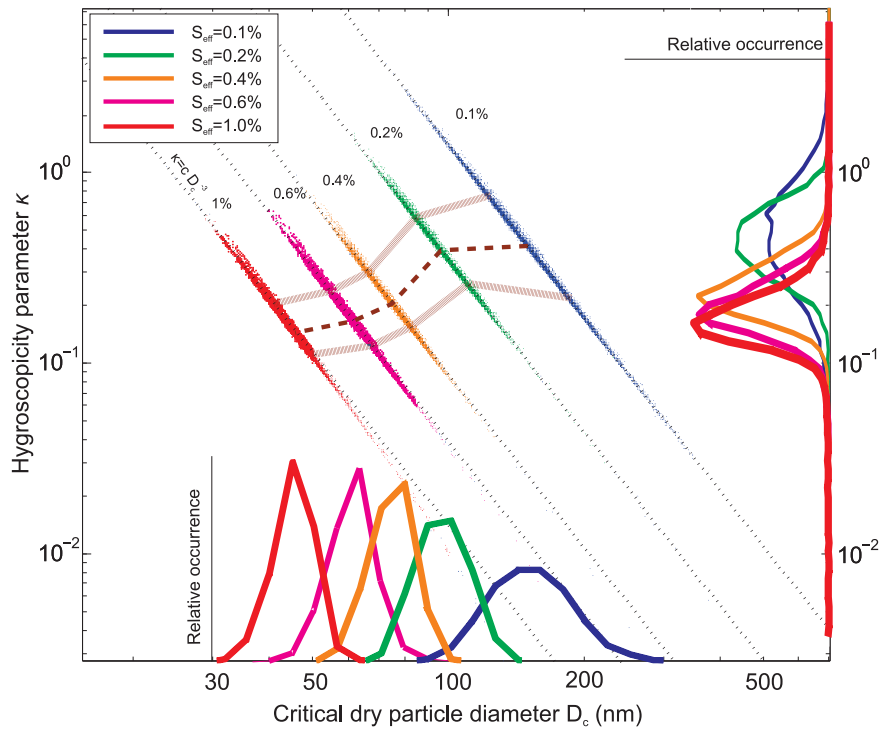Interactive comment on Atmos. Chem. Phys. Discuss., 13, 9681, 2013.

Fig. 1. Figure 7

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