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Atmospheric Chemistry and Physics Discussions

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 #2

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In the present paper Paramonov et al. have presented the long term measurements of cloud condensation nuclei using size-resolved technique. The measurements were carried out at in northern Finland at SMEAR II station in Hyytiälä over the period for more than two years. The dataset reported appears to be of high quality specially since it is a long term data set and I believe that manuscript adequately meets the standard of ACP. I, however, have following comments, which authors need to consider before manuscript is finally accepted in ACP.

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.

General comments:

I am not sure if the manuscript adequately addresses the message in the title. I believe it is more so important when no chemical data is presented and direct link (may be through modeling) related to clouds under given environment is presented.

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 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 throughout the paper.

1. I am not quite convinced with what authors have presented about Dc during NPF and non NPF days. I believe it is well expected that NPF would have some effects on Dc; having said that I think if authors could have had the chemical composition data would have been interesting to see during this period. In addition instead of Dc authors might consider showing κ for the relevant diameter during NPF and non-NPF events.

Response: Two previous studies about the hygroscopic properties of aerosol in

Hyytiälä reported that the difference in growth factors GF (Ehn et al. 2007) and Dc (Sihto et al. 2011) for particles of ∼50 nm in diameter between event and non-event days is small and inconclusive, which was also shown in the current manuscript. Considering the length of the dataset in the current manuscript and several subsets of event and non-event days analysed (page 9705, lines 12−16), the authors of this manuscript are fairly confident in the findings of the absence of clear effect of NPF on the hygroscopicity of small, ∼50 nm aerosol. It seems as though by the time the new formed particles grow to ∼50 nm in diameter, their CCN-relevant chemical composition is indistinguishable from that of the background aerosol. Considering that in all figures (e.g. Figs. 8 and 9) Dc and kappa mirror each other, the impact of plotting kappa instead of Dc in Fig. 10 is negligible. It was, in fact, done; however, the figure would show similar results with respect to the effect of NPF on kappa of ∼50 nm aerosol and, therefore, not present anything new.

2. As rightly pointed out by Referee#1 the average κ during Feb seems to be too high. Do authors believe that it has something to do with CCN calibration for correct effective supersaturation (too high supersaturation). This is more so important when the area under study is expected to be dominated by organics

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 ∼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. The paragraph in page 9702 has been altered to read: "It is plausible that the more active SOA formation and the increased organic fraction

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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 highest 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 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 comments:

1. Part 2, Theory in the manuscript may be shortened as reader may be a-priory familiar to the theory.

Response: The reader may or may not be a priori familiar with the theoretical background of this work. The authors believe that the theoretical framework is presented as concisely and comprehensively as possible, including all relevant information. Even though the theory part is, indeed, long, it is decided to leave it in its original form to make sure the reader gets a comprehensive theoretical overview and understanding of the background of this work.

2. There are some previous campaign based measurements from the similar location by other groups. Authors might want to compare the results; especially for that of hygroscopicity parameter.

Response: The three previous short-term campaigns in Hyytiälä by Ehn et al. (2007), Sihto et al. (2011) and Cerully et al. (2011) are all included and discussed in detail in the current manuscript. Other studies, such as Väkevä et al. (2002) and Wu et al. (2012), deal mostly with the hygroscopic properties of much smaller, CCN-irrelevant, nucleation mode particles. The authors of the current manuscript will gladly include any other relevant studies from the similar location suggested by the referee.

3. Figure 7 is relatively complicated to follow. I would suggest that authors split it and present it in much simpler way.

Response: The authors agree that the figure is indeed fairly complex; it does, however, present several important findings in a nice and condensed manner. In order to increase the clarity and simplicity of the figure, a legend has been added and the figure caption has been altered to read: "Figure 7. Distributions of κ values as a function of dry particle diameter. The different colours indicate different Seff levels, and the sides of the figure show relative occurrence of κ (vertical axis) and Dc (horizontal axis) calculated with log-equal bins. The dashed and the adjacent lines show the 25th, 50th and 75th percentiles of κ for each Seff level. The gray dotted lines show expected $\kappa \sim$ Dc$-$3 relationship for each Seff level."

4. I am not sure if authors wish to show Fig. 11 as it may be little confusing especially since it is difficult to explain why inactive fraction is negative in Jul. In addition in relation to Fig. 8b the highest kappa is in the month of Feb which is not consistent with lowest 1-MAF, which indicate something to do with supersaturation (calibration).

Response: The authors believe that Fig. 11 is an important figure and provides an insight into the seasonal variation of the aerosol mixing state in the boreal environment. This is especially true considering the total length of the dataset and the resulting degree of confidence of the resulting statistics. The authors agree that Fig. 11 is well explained in the text. Reasons for a negative CCN-inactive fraction in July are briefly mentioned in page 9706, lines 18$-$23 and elsewhere in the text where the poor statistics associated with very low particle concentrations at larger sizes are men-

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tioned (page 9694, lines 14–18, 20–22; page 9696, lines 10–12). As mentioned in page 9693, lines 4–19, the non-normalised method of fitting Eq. 2 to each activation spectrum excludes the CCN-inactive fraction from the calculation of Dc and kappa; and as mentioned in page 9696, lines 1–5, only the Dc and kappa from the non-normalised method are discussed throughout the paper. Therefore, the highest kappa in February has nothing to do with the CCN-inactive fraction, because this CCN-inactive fraction is not included in the calculation of kappa. In other words, what the authors think is happening in February is that ~5% of aerosol in 75–300 nm size range is completely non-hygroscopic (insoluble and refractory compounds) and these particles do not activate at any Seff within the CCNC; meanwhile the rest 95% of the aerosol of the same size happen to be very hygroscopic (for reasons discussed above). The figure caption has been revised to read: "Monthly median CCN-inactive fraction (1 – MAF), calculated from the non-normalised fitting of Eq. 2 to each activation spectrum. Error bars are 25th and 75th percentiles."

5. The diurnal plot of kappa and Dc for winter is not clear to me why these variations are like this; again if Feb has the highest kappa value it is not seen in the diurnal plots. Do authors have any explanation for it?

Response: The diurnal plots shown in Fig. 9 depict Dc and kappa for particles measured at the Seff of 1.0%, i.e. particles around 50 nm in diameter (Table 1).The highest kappa in February in Fig. 8b is for the Seff of 0.1%, i.e. particles around 150 nm in diameter. The winter plot in Fig. 9 shows kappa values ranging roughly between 0.14 and 0.18, which is also seen in Fig. 8b, where kappa for this supersaturation, shown as an empty red circle, is also around 0.18. Everything depicted in Fig. 9 is relevant only to particles measured at the highest Seff of 1.0%, i.e. particles of ~50 nm in diameter.

6. Some more discussion regarding the comparison between hygroscopicity parameter is required.

Response: This is a fairly unclear comment, and it does not point out as to where the suggested discussion should take place within the paper. Currently the paper includes an extensive comparison of the findings with several other studies, both at the same and in other locations around the world. The comment does not specify which kind of comparison is suggested, where in the manuscript the discussion is insufficient, and how the suggested addition would improve the quality of the paper.

References:

Asa-Awuku, A., Engelhart, G. J., Lee, B. H., Pandis, S. N., and Nenes, A.: Relating CCN activity, volatility, and droplet growth kinetics of Î2-caryophyllene secondary organic aerosol, Atmos. Chem. Phys., 9, 795-812, 2009. Bougiatioti, A., Fountoukis, C., Kalivitis, N., Pandis, S. N., Nenes, A., and Mihalopoulos, N.: Cloud condensation nuclei measurements in the eastern Mediterranean marine boundary layer: CCN closure and droplet growth kinetics, Atmos. Chem. Phys. Discuss., 9, 10303-10336, 2009. Gunthe, S. S., Rose, D., Su, H., Garland, R. M., Achtert, P., Nowak, A., Wiedensohler, A., Kuwata, M., Takegawa, N., Kondo, Y., Hu, M., Shao, M., Zhu, T., Andreae, M. O., and Pöschl, U.: Cloud condensation nuclei (CCN) from fresh and aged air pollution in the megacity region of Beijing, Atmos. Chem. Phys., 11, 11023-11039, 10.5194/acp-11-11023-2011, 2011. Su, H., Rose, D., Cheng, Y. F., Gunthe, S. S., Massling, A., Stock, M., Wiedensohler, A., Andreae, M. O., and Pöschl, U.: Technical Note: Hygroscopicity distribution concept for measurement data analysis and modeling of aerosol particle hygroscopicity and CCN activity, Atmos. Chem. Phys. Discuss., 10, 1005-1034, 2010.

Additional references used in the answer to the referee:

Väkevä, M., Kulmala, M., Stratmann, F., and Hämeri, K.: Field measurements of hygroscopic properties and state of mixing of nucleation mode particles, Atmos. Chem. Phys., 2, 55-66, 2002. Wu, Z., Birmili, W., Poulain, L., Merkel, M., Fahlbusch, B., van Pinxteren, D., Herrmann, H., and Wiedensohler, A.: Particle hygroscopicity during atmospheric new particle formation events: implications for the chemical species contributing to particle growth, Atmos. Chem. Phys. Discuss., 12, 11415-11443, 2012.

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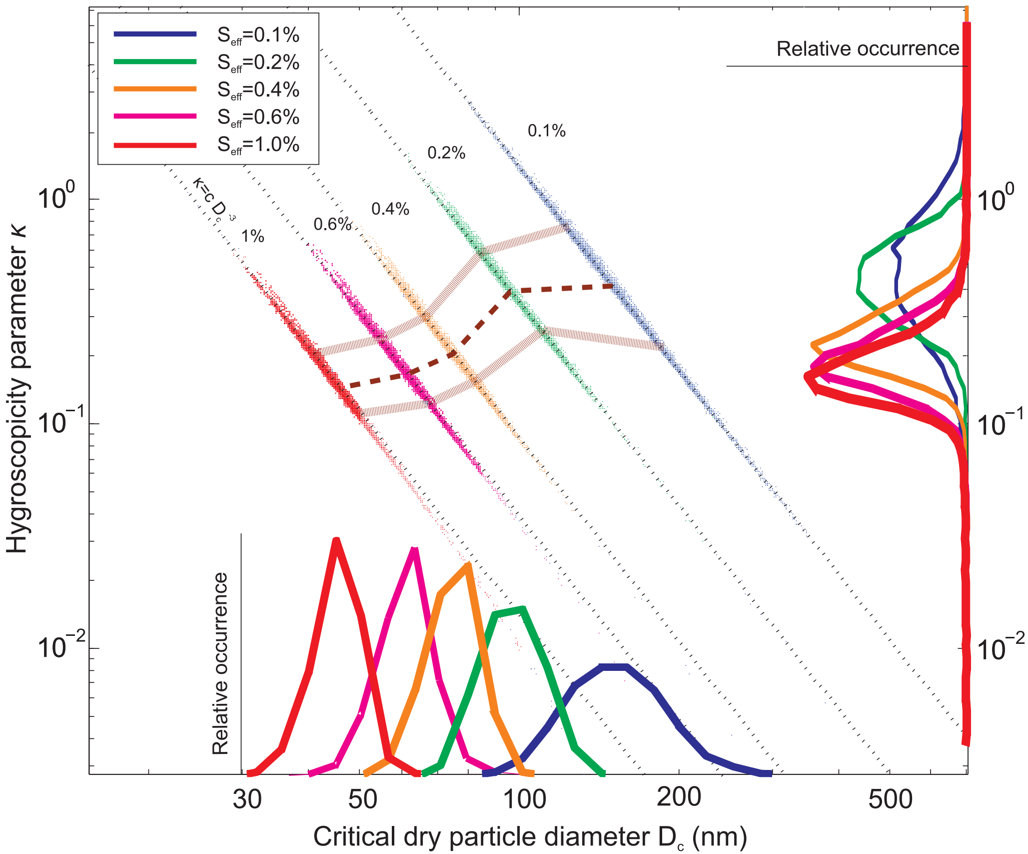

Fig. 1. Figure 7

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