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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measured and modelled cloud condensation nuclei concentration at the high alpine site Jungfraujoch" *by* Z. Jurányi et al.

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The authors would like to thank Anonymous Referee #1 for the review and for the constructive comments.

Below numbering refers to the referee's numbering

General comments:

1. Yes, the referee is right, the influence of chemical composition on CCN concentration is discussed in great detail while the influence of particle size distribution is just touched (Sect. 4.4 and Fig. 6). The reason for this is that the shape of the number size distribution is a highly variable aerosol parameter and with that a one month period is too short to make well-founded statements about its variability. However, in the mean time we

have 17 months of continuous CCN concentration and size distribution measurements. This data set will be used in an upcoming paper for a much more representative study about the influence of the variability of the size distribution on the CCN concentration. This is now emphasized in the revised conclusions section: "...The campaign mean kappa value was found to be $\sim$0.34. A detailed sensitivity analysis showed that this kappa value is in a range, where a variation in the kappa value around its mean value has only a small influence on the resulting predicted CCN concentration. Therefore it can be expected that the CCN concentration at the Jungfraujoch site can generally be well predicted from size distribution data with assuming a surface tension of pure water and a constant hygroscopicity parameter of $\sim$0.34. A continuous 17-months data set of parallel CCN number concentration and CN number size distribution measurements at the JFJ site has been acquired in the mean time. This data set will make it possible to provide a more representative value for the mean kappa value and to quantify the sensitivity of the CCN concentration to the variability of the aerosol number size distribution."

2. Comparison of the kappa values measured at the JFJ with kappa values at other sites around the globe is added to section 4.1: "This value is well representative for the global mean kappa values for continental regions (0.27 ± 0.21) whereas the mean value for marine regions is much higher (0.72 ± 0.24; Andreae and Rosenfeld , 2008; Pringle et al. , 2010)."

Discussion of the implications of the sensitivity analysis for other global sites has been extended in section 4.6: "From this analysis we can conclude that a change in the chemical composition has only a small influence on the predicted CCN concentrations except for very low SS. Since the campaign mean kappa value (0.34) is representative of continental sites around the globe (Andreae and Rosenfeld, 2008; Pringle, 2010), similarly weak sensitivity of CCN concentrations to chemical composition can also be expected for other continental sites. Even smaller sensitivity to chemical composition is expected for marine regions where the mean kappa is higher (0.72 ± 0.24). How-

ever, it is important to note that the sensitivity to chemical composition might be more pronounced in places, where the aerosol is much less hygroscopic with lower kappa values as for example reported in rainforests (Gunthe et al., 2009) or occasionally also at a site in Northern Europe (Kammermann et al., 2010a)."

3. The manuscript has been reworked with the aim of improving the clarity of this manuscript as requested by this and other referees. Specifically, the "results" and "discussion" sections have been merged into a single "Results and discussion" section. The latter has been divided into 8 subsections, each starting with an explanation of what is presented for better guidance of the reader. Additional information and interpretation has been added as requested with the specific comments.

Specific comments:

1. The paragraph is reformulated according to the request and reads now: "The critical supersaturation (SScrit), defined as the supersaturation (SS) at which the cloud droplet activation will take place, is mainly determined by the diameter of the particle at the point of the activation. This activation diameter depends on the dry diameter and the water uptake at RH below activation (hygroscopicity) of the aerosol particle (Köhler, 1936). The equilibrium vapour pressure over a curved pure water surface is elevated, which hinders the CCN activation. The process of activation can also be influenced by surface active species reducing the surface tension (Shulman et al., 1996; Shilling et al., 2007; King et al., 2009) and by compounds with limited solubility. . ."

2. The description of our work at the end of the introduction section has been expanded: "Here we present for the first time a CCN closure study covering a wide range of SS (0.12%–1.18%) from a remote continental measurement site which is most of the time situated in the free troposphere and only sometimes influenced by injections from the planetary boundary layer (Nyeki et al., 1998). Here we show that highly time resolved aerosol number size distribution data along with knowledge of the mean chemical composition is sufficient for accurate prediction of CCN number concentrations at

all investigated supersaturations. A detailed sensitivity study shows that the variability of chemical composition has little influence on CCN number concentrations, whereas the variability of the aerosol number size distribution has a bigger influence."

3. A brief description of the total inlet has been added: "Air was sampled through a heated (25 °C) inlet in order to evaporate any water that is associated with those aerosol particles that formed cloud droplets or ice crystals. The inlet has been designed so that it samples not only the interstitial aerosols but also the cloud droplets and ice crystals up to 40 μm. A detailed description of this "total aerosol inlet" is given in (Weingartner et al., 1999; Henning et al., 2002). Heating the aerosol from $\sim$-4.5 °C (mean outdoor temperature in May 2008) to $\sim$25 °C lab temperature also dries it to RH<10%.

4. A table containing instrument details has been added.

5. A reference to Köhler theory has been added.

6. The theory has been rearranged, reworded and additional equations are provided for clarification.

7. The following text has been added according to the referee's suggestion: "For our CCN closure experiment we measured the following aerosol properties throughout May 2008: CCN number concentration at different supersaturations as well as dry particle number size distribution and chemical composition. Time series of these measurements are shown in Figs. 1-3."

8. & 19. Yes, there is sometimes the mentioned relationship between AF and the CCN concentration in this data set, however, this is not always the case. For example between 26th and 28th of May the CCN concentration has increased a lot but the AF decreased or remained stable. We agree that such a potential relationship may be of importance and is worth deeper investigation. However, the continuous 17-months data set of parallel CCN number concentration and CN number size distribution mea-

surements, which has been acquired in the mean time, is much better suitable to address this issue rather than the limited data set of this study. The following sentence has been added: “Some correlation between the CCN number concentration and the activated fraction was observed for this data set (Fig. 1). Whether this is generally the case will be addressed in a forthcoming manuscript using a separate long-term data set.”

9. Interpretation of the variation of particle size distribution has been added: “A trend towards slightly bigger particle size during the period with influence from the planetary boundary layer compared to the free tropospheric conditions was observed. This is in line with the findings previously reported by Weingartner et al. (1999) based on a whole year of size distribution measurements.”

10. Figure 3b was already discussed in lines 3-6 on page 8871 of the ACPD paper. However, Figure 3c was erroneously referenced instead of Figure 3b. This has been corrected.

11. An interpretation of the results shown in Figs. 3c and 3d has been added: “The relative contribution of NH4NO3 and (NH4)2SO4 to the total inorganic volume fraction was changing significantly during the measurement period, whereas the inorganic to organic ratio exhibited much less variability. Since the kappa values of the different inorganic salts are similar, the overall kappa value does not experience large variations either (Fig. 3d). The campaign mean AMS/MAAP derived kappa value was 0.34. This value is well representative of continental regions (0.27 ± 0.21) whereas the kappa value in marine regions (0.72 ± 0.24) is much higher (Anreae and Rosenfeld , 2008; Pringle et al. , 2010).”

12. & 13. & 14. This paragraph has been rewritten for clarification: “CCN predictions of our closure study agreed well with measured values for every single data point, which is reflected in a fitted slope of nearly unity as well as a high correlation coefficient (Rˆ2>0.97) between the two quantities. In a different CCN closure study Lance et al.

(2009) reported agreement between prediction and measurement on average (fitted slope near unity), whereas the disagreement for individual data points was considerable (small correlation coefficient). The improved performance of our closure study is likely due to the fact that the number size distribution and chemical composition are more stable for the remote JFJ aerosol, which is not influenced by local pollution in contrast to measurements performed in the planetary boundary layer. Potential reasons for degrading performance of CCN closure studies conducted at sites closer to sources are: increased temporal variability of aerosol number concentration, shape of the number size distribution and chemical composition. In addition the assumption of size-independent chemical composition may become invalid (Ervens et al., 2009) and a substantial fraction of externally mixed particles with very low kappa values may be present.”

15. Text rearranged according to the referee’s request.

16. The comparison of HTDMA, AMS/MAAP, and CCNC derived kappa values has been rewritten and moved into a separate subsection (Sect. 4.7).

17. & 18. These two paragraphs have been moved into a separate subsection (Sect. 4.2) complemented by an introductory sentence.

20. SDE influence is now discussed in a separate subsection (Sect. 4.8).

Figures are modified according to the referee’s requests.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 8859, 2010.