Authors' responses to comments of Referee #1

We thank Referee #1 for the valuable comments for improvement of our manuscript, which we will implement upon revision. Detailed responses to the individual comments are given below. The referee's comments are listed first in italics, followed by our responses in normal font.

General comments:

RC: The authors do a good job of describing the measurements and results, but draw very few scientific conclusions from their data. Further, little explanation is given in the introduction for how these measurements add to or improve our understanding of aerosol hygroscopicity. For example, the authors state that the campaign averaged kappa value was similar to the generally assumed value for continental locations, 0.3, but then show that kappa actually changed significantly with aerosol size, air mass origin and weather events. Are these changes important or should modelers just use the campaign averaged value and not worry about these changes in hygroscopicity? Some discussion of the importance, or lack thereof, of these changes in kappa, perhaps discussing the importance of size resolved kappa representations in models or regional differences in kappa, would greatly strengthen the results of this paper and provide a more compelling motivation in the introduction.

AC: Together with the aerosol particle size distribution the hygroscopicity parameter kappa can be used to predict the total CCN concentration, which is an important input parameter for atmospheric cloud and climate models. The importance of kappa in the prediction of CCN concentrations was initially planned for a follow up paper. However, as both the referees requested a discussion of this topic for this paper, we performed a sensitivity study, which we plan to add in the revised manuscript. Briefly, the result of this sensitivity study is the following: For all supersaturations except the lowest level the assumption of a constant kappa value (campaign average of 0.31 or global average of 0.3) lead to a predictability of CCN concentrations within 20%. Using the bulk organic mass fractions actually measured by the AMS to calculate kappa did not lead to significantly improved predictability – the predicted concentrations differed still around 15-20% from the measured ones. Only the assumption of a size-dependent particle composition yielded further improvement in the CCN predictions, whereby the assumption of an campaign average size-dependent kappa value was not much worse than the size-dependent kappa derived from individual measurements of the organic mass fraction.

Generally the predictability of CCN concentrations for the lowest supersaturation level (0.079%) was much worse than for higher levels – the deviations between prediction and measurement were about twice as large. The reason for this difference could be found in the shape of the particle size distribution and the assumption of a purely homogeneous aerosol that was made in the calculation of the total CCN concentration (more details can be found in Fig. C1 below).

From this it results that for the prediction of CCN on a large-scale (e.g., in climate models) the assumption of an average kappa that is representative for this location (e.g. 0.3 for continental populated regions) is sufficient, whereas on a smaller scale (e.g., highly resolved cloud models) it is recommended to account for the size-dependent chemical composition to

calculate a more accurate kappa value. Additionally, the heterogeneity of the aerosol has to be approximated in particle size ranges for which the CN size distribution is rather steep. Note, however, that knowledge of the actual particle size distribution is much more important than knowledge of the particles' exact chemical composition (e.g., Dusek et al., 2006, Juranyi et al., 2010).

RC: The authors mention many other papers which present size resolved CCN and derived aerosol hygroscopicity but could do a better job of discussing their results in the context of these other studies. For exam.ple, Levin et al.(2013) performed very similar analysis comparing size resolved CCN measurements and AMS measured composition at a high altitude site. Their results are similar to those shown here with organic fractions increasing at smaller diameters, although overall their kappa values were lower. Paramonov et al. (2013) also found similar trends of aerosol composition and hygroscopicity with size. These, as well as other studies (Levin et al., 2012; Sihto et al., 2011; Gunthe et al., 2009), have also found background continental kappa values lower than 0.3 in areas far from human sources. The values of kappa shown in this work, therefore, seem to indicate some influence from anthropogenic sources. Some discussion of how the results presented here compare and differ from these previous studies and what, exactly, is represented by the measurements presented here (i.e. is this really a background site or representative of a mixture of natural and anthropogenic sources) would also greatly increase the utility of this work.
AC: In Sect. 3.2.2 we already mention that our findings compare well with earlier studies at the JFJ. Compared to the average kappa values from other continental background/rural sites in forested (mountain) regions (e.g., Fors et al., 2011; Levin et al., 2012, 2014; Paramonov et al., 2013) we observed rather higher kappa values, which can be attributed to differences in the observed organic mass fractions. Paramonov et al. (2013), who performed CCN measurements over one year, observed significantly higher kappa values during the winter season (kappa = 0.2 – 0.5 for particles with D = 40 – 150 nm) when probably much less organic material is emitted from the biosphere. These values agree very well with our observations at the JFJ in wintertime. As suggested by the referee, we will add this comparison in the revised manuscript. To avoid potential confusion or misunderstandings with regard to the meaning of "continental background site/station" we avoid this term in the revised manuscript and use the unambiguous term "high alpine site/station" instead.

Specific comments:

RC: Page 32579, Line 10: What is meant by "larger particles"? Give a specific size.
AC: With „larger particles" it is meant the accumulation mode particles with diameters of about 100 to 500 nm. We will add this information in the revised manuscript.

RC: "Page 32580, Line 20: Four distinct regions are identified from the HYSPLIT analysis, but only those from the easterly direction are discussed in the results. Some discussion should be included about air masses from these other directions, even if it is just to say that no changes in kappa or Nccn were detected. Have previous studies at this site seen any changes in aerosol type or concentration due to air mass origin?
AC: No significant differences in CCN parameters have been found for air masses from

northerly or (south-) westerly directions. Only the CCN parameters observed in air masses from easterly directions were distinctly different from those observed in the remaining time. We will clarify this finding upon revision of our manuscript. Also other studies from this site did not observe much variation with air mass origin.

RC: Page 32581, Line 10: What RH was achieved in the heated inlet?
AC: The relative humidity in the inlet was not measured but it can be calculated that even for a maximum outside temperature of -5°C and RH of 100% the sampled air should have a RH < 15% at an inside temperature of 25°C. Therefore the aerosol can be regarded as completely dry.

RC: Page 32585, Line 26: Could the SMPS measured size distribution be used to calculate losses?
AC: Yes, it can be used for loss calculations and indeed we did that in the comparison of NCCN,tot,m(MPI) and NCCN,tot,p(PSI) as described on page 32588, lines 5-9. However, we cannot apply a loss correction, which is size-dependent, to the directly measured NCCN,tot,p, for which no information on size is available.

RC: Page 32587, Line 2: Why was nitrate and not sulfate used to scale ammonium?
AC: The integrated mass concentration of ammonium was measured using the mass spectrum mode of the AMS. The duty cycle of the mass spectrum mode is by a factor of 50 higher than that of the size distribution mode, thus only the size distribution of ammonium was too noisy. For that reason the shape of the size distribution of ammonium was assumed to be equal to that of nitrate, since the inorganic species are usually internally mixed.

RC: Page 32588, Line 28: Were gravitational and inertial losses accounted for? Could these explain the differences?
AC: The line losses were determined by comparing the number concentration of size-selected aerosol particles upstream and downstream the sampling line using two CPCs. The applied correction should thus account for diffusional as well as gravitational and inertial losses. Upon revision of our manuscript we will delete the sentence.

RC: Page 32594, Line 26: An assumed aerosol density was used to convert AMS measured aerodynamic diameter to physical diameter, why not use the same assumed density here? Also, some mention should be made that using mass fractions instead of volume fractions will lead to an overestimate of kappa, since the density of the inorganic fraction is likely higher than that of the organic fraction.
AC: Test calculations using a density of 1.4 g cm-3 for organics and 1.8 g cm-3 for inorganics yielded relative differences less than 2% between kappa values calculated from a volume fraction-based parameterization and kappa values calculated from a mass fraction-based parameterization. For simplicity and consistency with related earlier studies we prefer to present the mass fraction-based parameterization directly related to AMS measurement data rather than introducing an additional/alternative volume fraction based parameterization. Following up on the referee's questions, however, we intend to address and clarify the relations between volume and mass fraction-based parameterizations in a follow-up study

presenting an overview of the correlations/parameterizations observed in various campaigns/locations.

RC: Page 32597, Line 19: More discussion should be included as to the differences between these measurements and those made previously at this site, as well as other sites as mentioned above.
AC: We agree with the referee and as already mentioned in the response to an above comment we will add more discussion upon revision of our manuscript.

RC: Figure 6: I suggest making this figure larger, or splitting it into multiple figures. As it is, it is hard to discern anything from it.
AC: We would like to keep the figure as is. Indeed it may be not easy to discern everything in it right now but with publication in ACP the figure would anyhow change to a more longish format, which should improve its readability.

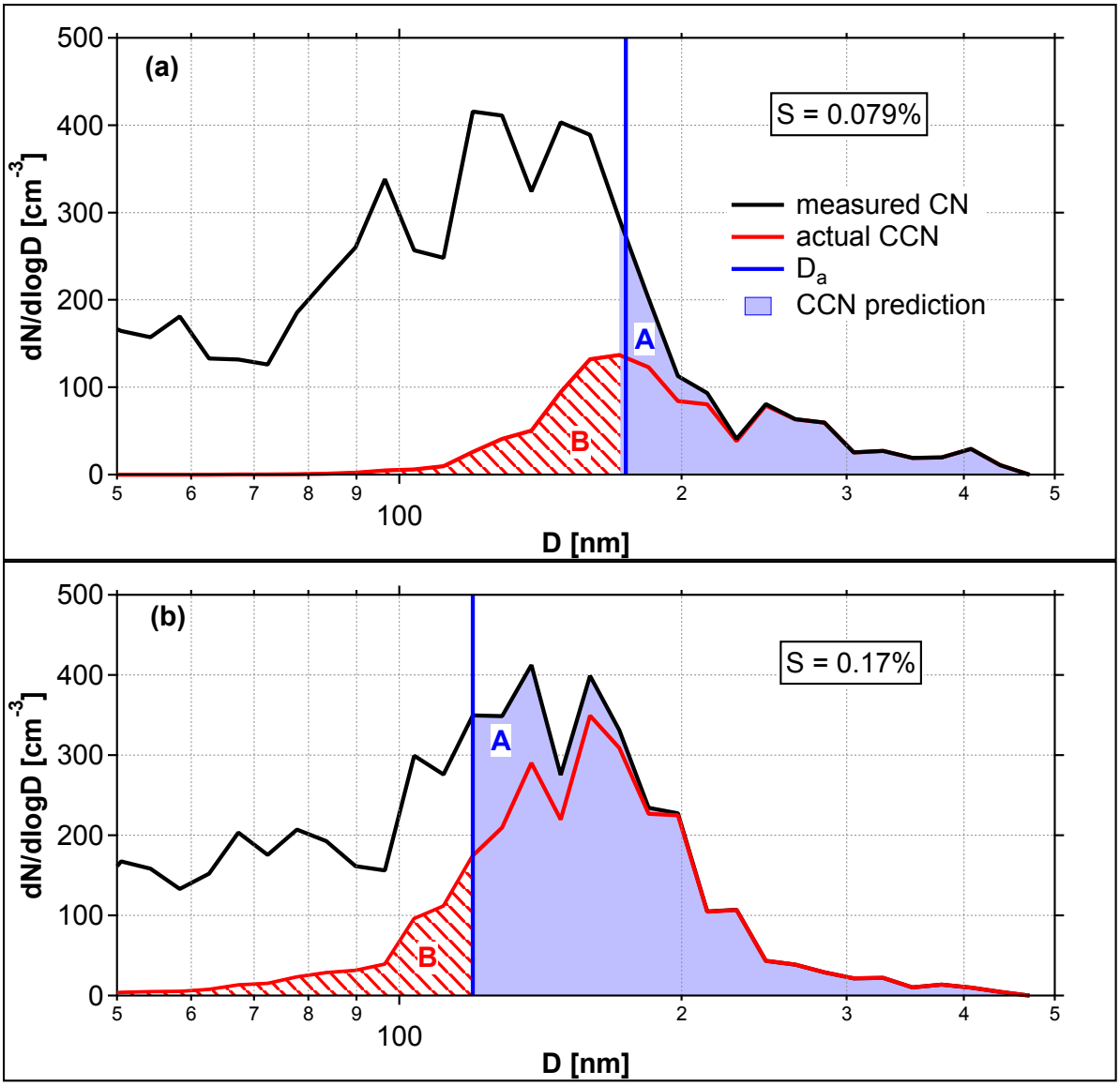

Figure C 1: Schematic sketch of how the total CCN number concentration is calculated exemplarily shown for two different supersaturation levels: (a) 0.079% and (b) 0.17%. The black line is the measured CN number size distribution and the red line is the actual CCN number size distribution calculated by multiplication of the CN number size distribution with the measured CCN efficiency spectrum (see Fig. 3a). Therefore the actual total CCN number concentration corresponds to the surface area below the red line. The predicted total CCN number concentration denotes the blue filled area, which corresponds to the surface area below the black line right of the activation diameter (indicated by the blue vertical line). The predicted CCN concentration matches the actual one only if the sub-areas A and B equal each other, which is the case for the larger supersaturations. For S = 0.079%, however, the shape of the CN size distribution and the heterogeneity of the aerosol leads to different sizes of the sub-areas A and B (A < B) and therefore to a significant underprediction of the CCN number concentration.