**Atmospheric
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Discussions

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Interactive comment on "Influence of cloud processing on CCN activation behaviour in the Thuringian Forest, Germany during HCCT-2010" *by* S. Henning et al.

S. Henning et al.

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We would like to thank both referees for the time invested in reviewing our manuscript. We highly appreciate their comments and hints for improving the paper. In the following we will address all comments and show how we changed the paper accordingly. We attached the changed manuscript text as pdf, where we highlighted the changes in the text in bold.

Answers to Anonymous Referee #1:

"The authors present two different methods for estimating the error in kappa, and each

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is based on different estimates of the uncertainty in set supersaturation. The first uses the accuracy in SS derived from repeated calibrations: presumably these were done as part of a separate study (Gysel and Stratmann, 2013), in which case what are the results of the calibration for the present study? The second method describes the error in SS as Gaussian with certain values of standard deviation, and applies Monte Carlo simulations. Could the authors explain where this assumption, and these values of standard deviation, comes from? It is not clear to me why the second approach is better than the first as the first uses an experimentally derived uncertainty in SS, and I would like the authors to please clarify this. As the authors point out, however, the main conclusion has already been tested by rigorous statistical methods, and the choice of error on kappa does not seem to affect this."

Our answer: You are correct; the calibration was not explained clearly enough and we have improved the explanation. The second method also uses SS values previously derived from repeated calibrations. The assumption of modelling the SS uncertainty by a Gaussian distribution is based on previous experimental results (repeated calibrations) showing that the error in instrumental SS values given by the CCNC is normally distributed. We have observed that the uncertainty of these instrumental SS values with 95 % confidence level is +/- 0.014 % for supersaturations $\leq$ 0.2 % and +/- 0.027 % for SS=0.4 %. These uncertainties are absolute values, i.e. SS = (0.1 +/- 0.014) %. According to the properties of the Gaussian distribution, 95 % confidence level corresponds to 1.96 standard deviations, in which case one standard deviation (σ) is 0.00714 % for SS $\leq$ 0.2 % and 0.01429 % for SS = 0.4 %.

As an answer to your second question why the second approach to estimate the error in kappa is better, we added the following paragraph in the text: Using maximum absolute error is a bad way of representing a Gaussian distribution, and since we know that the error in SS is Gaussian, the original error bars are a crude approximation. By assuming a Gaussian distributed SS error we are able to calculate the uncertainty distribution of kappa (by Monte Carlo sampling), and from this distribution it is easy to calculate

percentiles with which to represent error bars at desired confidence level. Percentiles, e.g. 95 % confidence intervals are a more correct way to represent the uncertainty in kappa than the maximum absolute error.

Changed manuscript text (second paragraph in section 3.1): In Fig. 3a and b the results are illustrated. The error bars were calculated by assuming a maximum absolute error in SS of ± 0.02% for SS =0.2% and assuming a 10% relative uncertainty for SS > 0.2% (Gysel and Stratmann, 2013), and applying Eq. (2) to calculate kappa. Due to the asymmetric nonlinear relation between SS and kappa also the error bars are asymmetric and give the maximum uncertainty in kappa. The increase in kappa after the cloud passage in the FCE is obvious, whereas in the NCE the data fall together on the 1 : 1 line. However, the observed effect is within the measurement uncertainty – especially for the lower supersaturations. Therefore, we tested the statistical significance of the change in critical diameters (and thus kappa values) between the stations during FCE and NCE, and re-estimated the uncertainty of kappa by modeling the instrumental error in supersaturation by a Gaussian distribution.

Changed manuscript text (third paragraph in section 3.2): Next, we estimated the uncertainty distribution of κ with Monte Carlo simulations. We have previously observed that the instrumental supersaturation error of the CCNc is Gaussian, with standard deviations of 0.00714 for 0.07 %, 0.1 % and 0.2 % supersaturations and 0.01429 for 0.4 % supersaturation. These standard deviations are obtained from repeated calibration results showing that with 95 % confidence level the absolute uncertainty for supersaturations $\leq$ 0.2 % is +/- 0.014 % and for SS = 0.4 % the uncertainty is 0.027 %. The 95 % confidence level corresponds to 1.96σ, from which we can derive the aforementioned standard deviations. However, due to the nonlinear relationship between κ and the critical diameter, the uncertainty distribution of κ is non-Gaussian. The distribution of κ is simulated for each data point separately by drawing 100 000 random samples from a Gaussian supersaturation distribution (μ = 0.07, σ = 0.00714) and using Eq. (2). An example of a simulated κ distribution is presented in Fig. 5, showing the 2.5, 25, 50,

75, 97.5 and 100th percentiles. All the analyses were done using R statistical software (R version 2.15.3, 2013).

By applying this statistical approach to the data, it is possible to present more realistic error bars. Using maximum absolute error is a bad way of representing a Gaussian distribution, and since we know that the error in SS is Gaussian, the original error bars are a crude approximation. By assuming a Gaussian distributed SS error we are able to calculate the uncertainty distribution of kappa (by Monte Carlo sampling), and from this distribution it is easy to calculate percentiles with which to represent error bars at desired confidence level. Percentiles, e.g. 95 % confidence intervals are a more correct way to represent the uncertainty in kappa than the maximum absolute error. Figure 6a gives single κ values at the upwind station compared to the κ at the downwind station during FCE. The error bars presented in the figure are the 95 % confidence intervals calculated from Monte Carlo simulations as explained above. All κ values derived for the downwind station are higher than those at the upwind station. The same analysis was again done for the NCE periods (Fig. 6b).

"In section 3.3, the authors state: "This estimate is supported by measurements results from other groups during HCCT-2010, who focused on the chemical and isotopic signature of the particle population". Can the authors please elaborate on this and provide a reference (if available). Is this the "personal communications" referenced later in this section? The conclusions of this section are supported by these results, so it would be useful, if possible, to provide some numbers / figures. I appreciate the data belong to other research groups, so their inclusion may not be feasible, but I would like at least to see some better referencing, and an elaboration of what these results are."

Our answer: The sentence "This estimate is supported by measurements results from other groups during HCCT-2010, who focussed on the chemical and isotopic sig-nature of the particle population". is referencing to the following paragraphs. Up to date,we cannot give a better referencing for the AMS measurements than personal communication / papers in preparation, because the findings are not yet published. The find-

ings based on stable isotope fractionation are meanwhile published in ACP and are referenced correctly. We rearranged the paragraph, which hopefully clarifies that the sentence was meant to be an opening statement for the following paragraphs.

Changed manuscript text: This estimate is supported by measurement results from other groups during HCCT-2010, who focused on the chemical and isotopic signature of the particle population; for example, sulfur isotope analysis of the particulate material was used to investigate the in-cloud production of sulfate. Combined gas phase and single particle measurements allowed the dominating sulfate production sources to be identified (Harris et al., 2014). Direct sulfate uptake, through dissolution of H2SO4 gas and scavenging of ultrafine particulate, was found to be the most important source for in-cloud addition of sulfate to mixed particles (the most common particle type at HCCT-2010), while in-cloud aqueous oxidation of SO2 primarily catalyzed by transition metal ions (Harris et al., 2013b) was most important for coarse mineral dust. The isotopic analyses showed that the sulfate content of particles increased following cloud processing at HCCT-2010 by >10-40% depending on particle type (cf. table 5 in (Harris et al., 2014)).

"Minor corrections: Page 1620, lines 24-26: "were achieved" appears twice in this sentence. Page 1623, line 12: Remove either "the" or "another"." The above mentioned typos were corrected.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C2849/2014/acpd-14-C2849-2014-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 1617, 2014.

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