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Interactive Comment

Interactive comment on "Assessment of cloud supersaturation by aerosol particle and cloud condensation nuclei (CCN) measurements" *by* M. L. Krüger et al.

Anonymous Referee #1

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Summary

"Assessment of cloud supersaturation by aerosol particle and cloud condensation nuclei (CCN) measurements" by Krüger et al. uses *in situ* scanning mobility particle sizer (SMPS) and size-resolved CCN data to estimate the water vapor supersaturation (S) of a cloud. This question is of interest to the atmospheric community, and the manuscript clearly and effectively communicates the techniques used and the theory that underlies them. Although many similar studies estimating in-cloud S have been published previously, none to my knowledge use CCN data for size-selected ambient particles as described in this manuscript. The authors show that estimates of S are consistent

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with those using more established techniques, although given the large uncertainties associated with the new estimates this is not at all surprising. Unfortunately, there are major issues with the values reported using the new method (i.e., the bottom row of Table 2) which need to be addressed. They are simply arithmetic averages of a subset of the five S_{CCNC} values at which the CCN instrument was operated. It is not clear if these averages have any physical significance, how dependent these values are on the somewhat arbitrary choice of temperature gradients used in the instrument (which determine S_{CCNC}), and in one case (S_{avg}) if the subset of S_{CCNC} values used is appropriate. I recommend publication in Atmospheric Measurement Techniques if the following issues can be addressed, although I think it is likely that more values of S_{CCNC} will be required to make this a viable technique.

Major comments

I would recommend that the authors change the title slightly to reflect what is truly novel about their analysis, which is the use of size-resolved CCN measurements to estimate cloud S. I suggest "Estimation of cloud supersaturation by aerosol particle and size-resolved cloud condensation nuclei (CCN) measurements."

Given that the major goal was to determine S in a cloud, a weakness in this work is that CCN data from only five values of S_{CCNC} are presented. Thus the reported ranges in S_{low}, S_{high}, and S_{avg} were very wide (0.19 – 0.25%, 0.90 – 1.64%, and 0.38 – 0.84%, respectively), and as discussed below the reported range in S_{low} should be greater. The authors state this on pg. 10037, ln. 6-10 by pointing out that use of a greater number of S_{CCNC} would "increase the precision of the derived cloud peak supersaturations." If the authors were to use this technique with scanning flow CCN analysis (Moore & Nenes, 2009), as they recommend for future work in the conclusions, this paper would be greatly strengthened.

pg. 10022, ln. 13-14: The value of S_{low} given is 0.19 – 0.25%. However, this is based on observations of no significant in-cloud CCN activation at S_{CCNC} = 0.13%,

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in-cloud CCN activation similar to out-of-cloud at S_{CCNC} = 0.25%, and no intermediate observations. The reported values of S_{low} should not be any more precise than the S_{CCNC} intervals, and therefore the range of S_{low} should be changed to 0.13 – 0.25%. In other words, the authors cannot say that the minimum S in the cloud was not lower than 0.19% without observations at that S_{CCNC}. The authors state on pg. 10030, ln. 21-23 that they use 0.19% because it is the mean value between the two closest observed S_{CCNC} levels, but it is not clear that this mean value has any real significance.

On pg. 10025, ln. 19-20, the authors state that "the time directly after the cloud event, when there was clearly no cloud present at the inlet, is referred to as 'out-of-cloud' conditions". Does this mean that out-of-cloud conditions were identified by increased ambient visibility? In other words, what do the authors mean by "clearly?" The authors then state that during in-cloud conditions, the LWC was > 0.024 g m^{-3} for 85% of the time. What was this percentage for out-of-cloud conditions? This distinction between in-could and out-of-cloud conditions is critical to the analysis, and so more detail is needed.

I recommend adding a figure with a time series of LWC during and just after the single analyzed cloud event. This figure would also support the caption for Fig. 3, which states that "averaging times were chosen to be unambiguous with respect to LWC for in-cloud (mean LWC = 0.131 g m^{-3}) and out-of-cloud conditions (mean LWC = 0.016 g m^{-3}) within a short time interval (in-cloud: 19 September 2012 15:00–16:00 UTC; out-of-cloud: 19 September 2012 17:30–18:00 UTC)." A time series of LWC from, e.g., 14:00 to 19:00 UTC would be highly relevant and would strengthen the manuscript.

On pg. 10032, ln. 18-21, the authors state that the "shoulder" at 40 nm in Fig. 4 "is likely due to aging processes such as condensational growth or coagulation, which are usually more pronounced for the Aitken mode than for the accumulation mode." While this is true generally, it seems unlikely that these aging processes would be present for in-cloud aerosol and absent for out-of-cloud aerosol measured about 1 hour later. It seems much more likely that some of the Aiken mode particles are activating in the cloud. It would therefore appear that some particles are activating at S greater than 0.68% (i.e., the maximum value of S_{CCNC} reported), as indicated by the derivation of S_{high} in Section 3.2.4.

The choice by the authors to neglect the shoulder in Fig. 4 is especially problematic given that their analysis assumes that the dry particle size distribution is the same both in-cloud and out-of-cloud. Beginning on pg. 10031, ln. 25, the authors state that "we had no opportunity to measure total aerosol properties under in-cloud conditions. For the investigated cloud event, however, the in-cloud and out-of-cloud measurement periods immediately followed each other without apparent changes in the regional atmospheric conditions. Thus, we assumed the total aerosol properties measured out-of-cloud to be approximately representative for the total aerosol properties in-cloud." Later, on pg. 10033, ln. 13-16, the authors state that "it was not possible to measure the particle size distribution of both interstitial and total aerosol inside the investigated cloud. As outlined above, however, we have good reasons to assume that the total aerosol size distribution measured out-of-cloud was approximately representative for the total aerosol in-cloud." These assumptions contradict the justification given for neglecting the shoulder in Fig. 4, which was essentially that the in-cloud aerosol was more aged than the out-of-cloud aerosol.

On pg. 10033, ln. 1, the authors report that the "average peak supersaturation" of the cloud was 0.48 $\pm$ 0.10%. It is not clear what is meant by "average" in this context – the authors state that S_{avg} is the point "at which most particles have been activated", but they have not demonstrated that, e.g., only at this point are > 50% of the particles CCN active. This value of 0.48% is simply the mean value of "three neighboring supersaturation levels", which are actually the three highest of the five S_{CCNC} used. I have a hard time attaching any significance to this value, mainly because it depends on the arbitrary choice of the five S_{CCNC} levels used in the CCNC, and also because it is not clear from Fig. 4 that all three provide equally good matches to the activated particle size distribution. If anything, the data from S_{CCNC} = 0.68 and 0.51% fit much better than those from S_{CCNC} = 0.25%. I don't think the authors can report an "average peak supersaturation" of the cloud using this method. Given these issues and the fact that data for particles smaller than 70 nm were neglected without sufficient justification (see above), the authors should remove section 3.1.2 from the manuscript.

If the authors wish to retain section 3.1.2 and the results described therein, I think at a minimum they need (1) a more clear definition of "average peak supersaturation," (2) to redo the analysis without neglecting particles smaller than 70 nm, (3) to find a way to estimate S_{avg} based on a more nuanced analysis instead of simply taking the arithmetic average of the three highest S_{CCNC} levels, and (4) to consider the possibility that the peak in-cloud S may at times be greater than the maximum value of S_{CCNC}. The authors allow for this possibility in section 3.2.4, when they derive S_{high} using established techniques based on SMPS data. Given that the main focus of this AMTD manuscript is on the novel approach of using size-selected CCN data to estimate S, however, this possibility should also be discussed in any revised section 3.1.2.

Minor Comments

On pg. 10035, ln. 11, the authors report a range of S_{avg} of 0.38 to 0.70%, based on the Hoppel minimum and several assumptions regarding particle hygroscopicity. They then report the mean value of 0.54%, with a standard error of 0.06%. But the standard error does not seem to be the best indicator of uncertainty here. The main source of uncertainty is probably the value of kappa (i.e. the hygroscopicity), not the counting statistics and other experimental parameters used to determine the standard error. I therefore recommend replacing the standard error in this case (and in Table 2) with the standard deviation of the S_{avg} values, i.e., $0.54 \pm 0.14\%$.

It is not clear why different times are used for in-cloud and out-of-cloud conditions in Figs. 1 - 2 and Fig. 3 (with Fig. 3 using more limited time intervals). The caption for Fig. 3 states that this was "to assure comparability of the size distributions", but it is not clear why this would not apply to Figs. 1 and 2. Please explain why more limited

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time intervals were used for Fig. 3, perhaps by making reference to the new LWC time series figure suggested above.

References

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Interactive comment on Atmos. Meas. Tech. Discuss., 6, 10021, 2013.

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