April 30, 2014

Response to the Referee #1 (Krüger et al., amt-2013-246):

We thank Referee #1 for the review and the constructive suggestions for improvement of our manuscript, which will be implemented upon revision. Detailed responses to the individual comments are given below. The referee's comments are listed first, followed by our responses:

Referee comment:
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."

Author Response:
The title has been changed according to the referee's suggestion to "Assessment of cloud supersaturation by aerosol particle and size-resolved cloud condensation nuclei (CCN) measurements."

Referee comment:
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.

Author Response:
The initial purpose of this measurement campaign was not to measure the cloud supersaturation. Therefore we used our standard measurement routine scanning 10 different particle diameters at five fixed supersaturation levels. In the end of the campaign we realized that with our setup it should be possible to estimate the cloud supersaturation. The actual data analysis we started only after the campaign. If we had had the intention to measure the cloud supersaturation before, we would indeed have set the CCNC to measure at a much larger number of supersaturation levels. However, we are not able to repeat the measurements at that site.

"The method was developed and applied for the investigation of a cloud event during the ACRIDICON-Zugspitze campaign (17 Sep to 4 Oct 2012) at the high-alpine research station Schneefernerhaus (German Alps, 2650 m asl)."

Will be changed to:

"The method was developed and applied during the ACRIDICON-Zugspitze campaign (17 Sep to 4 Oct 2012) at the high-alpine research station Schneefernerhaus (German Alps, 2650 m asl).

The intention of this manuscript is to present the approach and to illustrate that it is in principle possible to measure S_{cloud} with this setup. We did not claim that the numbers that we got for S_{cloud} are necessarily exact values. That is the reason why we provide a quite large range of S_{cloud}. In the revised version of the manuscript we will add another sentence to make clear that this study is only a proof of principle.

The following text will be added to Sec.4 of the revised version of the paper:

"This study, however, mainly presents an alternative method to estimate the cloud supersaturation. Thus, the numbers that we got for S_{cloud} are not necessarily representive values."

<u>Referee comment:</u>
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%, 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.

<u>Author Response:</u>
In the abstract S_{low} was given as 0.19% to 0.25%. This range was based on Table 2 (second column) and includes the range of values for S_{low} obtained by different methods (SMPS and CCNC). In Table 2 the value for S_{low} we obtained from the CCNC method during one single cloud event was given as 0.19% +/- 0.04 (mean value between highest level of no activation (0.13%) and lowest level of activation (0.25%) +/- standard error between these two values).
We agree that it is not meaningful to use the standard error to express the range of possible S_{low} values. Instead it is more correct to include the full range of possible S_{low} values. In our case S_{low} must be somewhere between *S*=0.13% and 0.25%. Therefore, in the revised version of our manuscript we will equivalently talk of 0.19% +/- 0.06%. In the abstract, however, we follow the suggestion of the referee and will change the statement into "For the investigated cloud event, we derived S_{low} ~ 0.07%-0.25%...", which includes the full range of S_{low} values we obtained from both the SMPS and CCNC method.

<u>Referee comment:</u>
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.

<u>Author response:</u>
The LWC exhibited high fluctuations. These short periods (on the order of a few minutes) of significant change in LWC, however, did not seem to change aerosol properties (e.g. number size

distribution) noticeably. For our analysis, we therefore introduced the criterion defining a period of CCN measurements as in-cloud when the LWC was > 0.02 g m^{-3} for 85% of the time and as out-of-cloud when the LWC was < 0.02 g m^{-3} for 85% of the time.
To avoid misunderstanding we will change the paragraph on pg. 10025, ln. 14-24 as follows: To distinguish between in-cloud and out-of-cloud conditions, we utilized measurements of the liquid water content (LWC), which were performed by a particle volume monitor (Gerber, 1991). As suggested also by Henning et al. (2002) we defined a period of CCN measurements as in-cloud, when the LWC was > 0.02 g m^{-3} for 85% of the time and as out-of-cloud when the LWC was < 0.02 g m^{-3} for 85% of the time. For the analysis in this paper we chose one exemplary cloud event, which occurred on 19 September 2012. During this event the LWC was on average 0.073 g m^{-3}.

Referee comment:
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.

Author Response:
A figure of the LWC will be added to the revised manuscript (Figure C1).

Referee comment:
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.

Author response:
The aging processes such as condensational growth or coagulation do not necessarily happen in cloud but mainly after the cloud has disappeared. Therefore we argued in the text that the Aitken mode particles grew leading to the shoulder in the size distribution of activated particles. Nevertheless it might be true that some of the Aitken mode particles were activated in the cloud. Upon revision of our manuscript we will change the paragraph as follows: "The number size distribution of activated particles exhibited a large peak at ~124 nm with a maximum of ~1000 cm^{-3} (dN/dlogD) and a shoulder at ~40 nm and ~350 cm^{-3} (dN/dlogD), whereby the shoulder is mainly in the range of the error bars. On one hand it may result from a slight shift of the CN size distribution between in-cloud and out-of-cloud conditions, which 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. On the other hand it may result from a locally higher supersaturation during the cloud event (S_{high})."

Referee comment:
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.

Author response:
The out-of-cloud aerosol was more aged than the in-cloud aerosol, not the other way around. The reason for this assumption was discussed in the manuscript text before. As discussed in the response right above, the shoulder is in the range of the error bars, which we will include in the figure (Fig. C2) upon revision. As suggested by the referee we will not neglect the shoulder but we also do not put too much emphasis on it.

Referee comment:
On pg. 10033, ln. 1, the authors report that the “average peak supersaturation” of the cloud was 0.48 +/- 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.

Author response:
As stated in the manuscript the average peak supersaturation (S_{avg}) is the supersaturation at which most of particles have been activated and formed cloud droplets. Here, the term “average” is not the arithmetic mean value or median value (or any other mathematically meaningful value) but a general term to express that there might be a variety of supersaturations that the aerosol particles experience.
The definition of S_{avg} determined by the CCNC method is the supersaturation level in the CCNC instrument at which the distance between the measured CCN size distribution and the size distribution of activated particles is smallest. In the revised version of our manuscript we will explicitly state this definition. So far the definition was mentioned only implicitly in the text (“Consequently, the number size distribution of activated particles in a cloud should be approximately equal to the CCN size distribution measured with the CCNC for total aerosol at a supersaturation level equivalent to the effective average peak supersaturation in the cloud“).
S_{avg} determined by the SMPS method is defined as the supersaturation that corresponds to the diameter at which 50% of the particles of this size are activated.
S_{avg}(CCNC) and S_{avg}(SMPS) are equivalent parameters but since they are defined differently the resulting values may be not exact same.

Referee comment:

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 of 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.

Author response:

We agree with the referee that it is not meaningful to take the mean value of the three neighboring supersaturation levels in this case. As mentioned in the above comment S_{avg}(CCNC) is the supersaturation level in the CCNC instrument at which the distance between the measured CCN size distribution and the size distribution of activated particles is smallest.

Upon revision we will also include the range of errors for the data displayed in Fig. 5. According to the above definition we take S = 0.51% +/- 0.06% as an estimate for S_{avg}(CCNC).

Referee comment:

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.

Author response:

We would like to keep Sect. 3.1.2 and will implement all referee suggestions as already discussed in the above comments.

Referee comment:

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+-0.14%.

Author Response:

Thank you for pointing this out. In the revised manuscript we will indicate the standard deviation instead of the standard error.

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

Author response:
The main reason for using different time periods in Fig. 1+2 and 3 is that different time is needed to perform one full spectrum with the two instruments (~2 hours for 5 S-levels with the CCNC and ~2 min for one particle size distribution with the SMPS). Due to the different measurement intervals it was necessary to shift the measurement times in the different figures. Nevertheless this shift was not optimal, so we adjusted the times slightly in the revised version.

Figures:

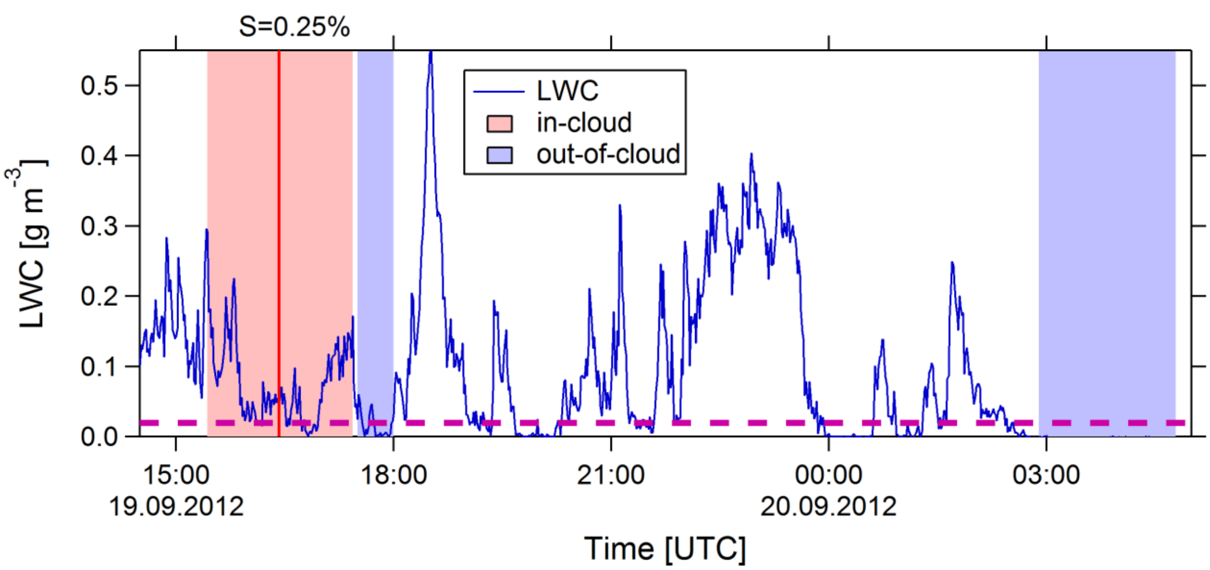

Figure C1: The time period for in-cloud conditions when the cloud supersaturation was investigated is marked read (19 Sep. 2012 15:26-17:17 UTC), The red line marked the time until there are information about the supersaturation by using the interstitial CCN data, for that reasoned for the SMPS method the data was taken until this time (19 Sep. 2012 16:25 UTC). The out-of-cloud conditions are market blue. The first small time period for out of cloud conditions was used as out-of-cloud condition using the SMPS method (19 Sep. 2012 17:30-18:00 UTC). For the size resolved CCN measurements it need more time to gut a complead scan, for that reasoned the data for out-of-cloud conditions using the CCNC method was taken later (20 Sep. 2012 02:54-04:47 UTC).

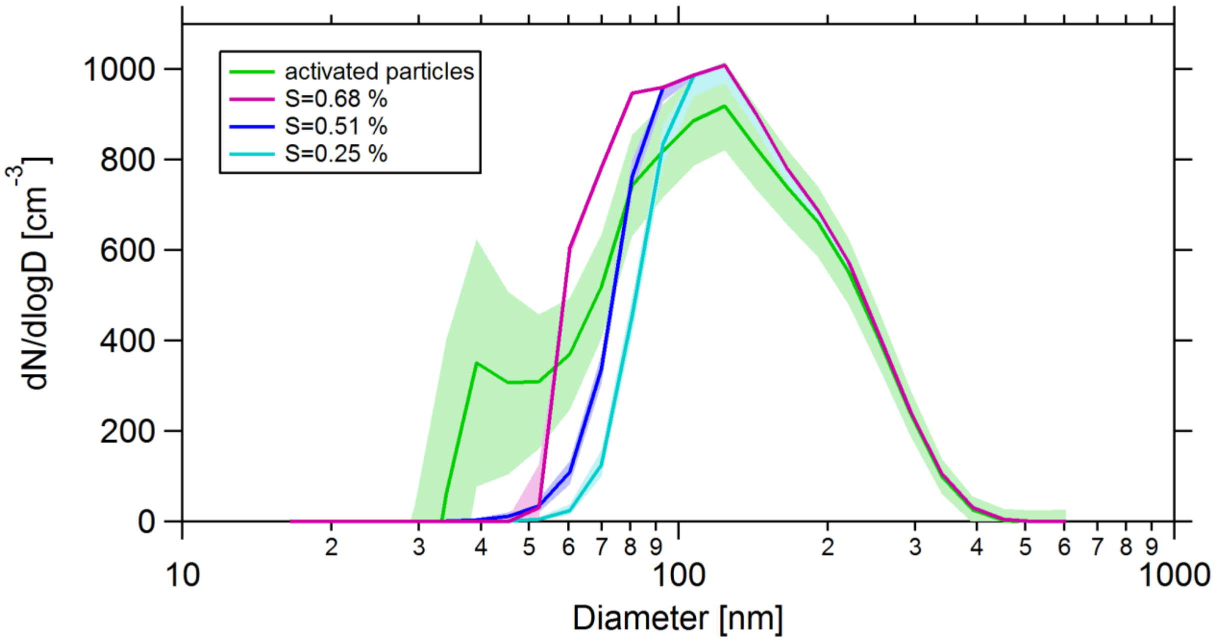

Figure C2: Average number size distribution of activated particles in the cloud (green; shaded area is the range of the statistical error of the data points) and CCN size distributions at S_{CCNC} = 0.25 %, 0.51 %, and 0.68% (colored lines; shaded area is the range of the statistical error of the data points).