April 30, 2014

Response to the referee #2:

We thank the referee #2 for her/his positive review. The minor revisions suggested by the referee have been useful to improve the manuscript quality and have been processed, as outlined in detail below. The referees' comments are listed first, followed by our responses:

Referee comment:
As far as I understand, the goal is to develop methods to measure in-cloud, in-situ peak supersaturations (or RH). The methods described are attempts to that, except the "Hoppel minimum" method (sect 3.2.2). That method is more a measurement of the history of the aerosol, i.e. the supersaturation that the aerosol was exposed to in previous cloud passages. Results from the "Hoppel minimum" method can anyway be interesting for comparison, but I cannot see that as a relevant method for in-situ measurements. Or maybe I misunderstood something here.

Author Response:
We agree with the referee that the Hoppel minimum tells more about the history of the aerosol instead of the actual conditions. In general methods using SMPS data do not allow to distinguish between particles, which have been activated at different supersaturations at a given time and location. In contrast, the CCNC-based approach to estimate S_{low} in this study provides measurements of the actual supersaturation at the instrument's inlet.

Referee comment:
The spread in results, see Table 2, seem rather large. The authors state in section 4 "Conclusions and outlook" that the uncertainties mostly depend on limitations in time resolution and counting statistics, as well as uncertainties in the aerosol hygroscopic properties for the SMPS methods. It might be interesting to also have a discussion and draw conclusions about the reliability of the different methods. Are all methods equally accurate?

Author Response:
With the small data set the study is based on it was not possible to redo the analysis for the SMPS and the CCNC method for another cloud air parcel. However, we had the chance to investigate the two methods seperately at some more examples, which shows a nice reliability for each of them. In the revised version of the manuscript we will improve the way of analyzing the SMPS data, which reduces the error bars (Fig. C1). This affects the results for S_{low} significantly (Table C1, C2), because the definition of S_{low}(SMPS) strongly depends on uncertainties of the SMPS measurement.

We will discuss this topic in Section 3.2.3 of the revised paper as follows:

"This range is not consistent with the estimate derived from the CCN approach (S_{low}(CCNC) = 0.19% ± 0.06%). A reason could be that the definition of S_{low} from the SMPS method strongly depends on uncertainties of the SMPS measurement. On the other hand, the SMPS approach tells more about the cloud history and not the about the actual supersaturation at the inlet, which is accessible by the CCNC method. Therefore, the discrepancy can be traced back to the fact that SMPS and CCNC methods measure the supersaturation from different time."

Referee comment:
The authors discuss already in the abstract the variability of supersaturations that the aerosol particles and cloud droplets are exposed to during the evolution of the cloud. How does this affect the results? Is it only a time variation?

Author Response:
As already discussed in the response above the methods are sensible to the variability of supersaturations. The SMPS method and the S_{avg} calculated by the CCNC method can mirror the history of the aerosol particles but cannot tell anything about the time the aerosol was activated into cloud droplets. S_{low} calculated by the CCNC method always gives the lowest supersaturation all particles have been exposed to, which is the supersaturation at the inlet for the case activation happens at the inlet.

Referee comment:
The section starting on page 10024, line 27, reaching until page 10025, line 3 (Most likely …) is most likely correct, but seem not very relevant to this paper. The techniques presented aim to measure supersaturation in a specific cloud at a specific time and location, i.e. the location where the instrumentation (inlet) is placed. Earlier cloud passages by the aerosol particles seem not very relevant to that parameter.

Author response:
We regard this statement as very relevant for the paper because it underlines the complexity and variability of the supersaturation of a cloud.

Referee comment:
I believe section 4 "Conclusions and outlook" could be improved, especially the conclusion part. It would be interesting if you could present a few more conclusions from your work, see also above.

Author Response:
The following paragraph will be added to section 4:

"The lower bound of cloud peak supersaturation (S_{low}) calculated by the CCNC method is significantly higher than S_{low}, calculated by the SMPS method. The following two effects may explain this discrepancy: (i) SMPS data analysis requires integration over a certain time period to reduce uncertainties. This has influence on the error, which is particularly important for the S_{low} estimation. (ii) All supersaturation estimates based on the SMPS method reflect the history of supersaturation, experienced by the aerosol particles during cloud evolution. Thus, particles, which have been activated once into cloud droplets, will be counted independent of the time when the activation actually occurred. If particle activation takes place during the measurements, the CCNC method provides in-situ measurements of the actual supersaturation in the probed cloud air parcel."

Referee comment:
Page 10023, line 11 and page 10046, line 23: Is "Pruppacher and Klett, 2010" really correct? My version is from 1997.

Author Response:
We referenced an online version of this book, which is from 2010.

Figures:

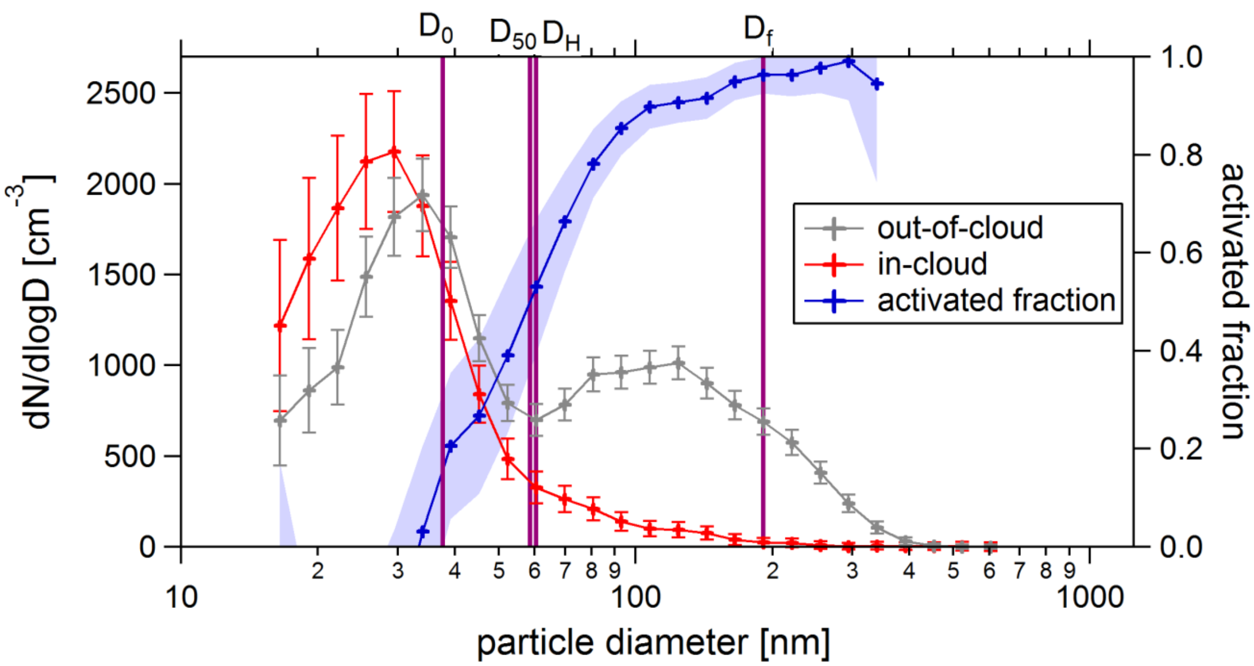

Figure C1: Average number size distribution of total aerosol particles (grey crosses; out-of-cloud) and of interstitial aerosol particles (red crosses; in-cloud). The error bars correspond to the individual standard errors calculated as described in Sect. 2.4. The activated fraction (blue crosses; grey minus red, divided by grey; shaded area is the range of the statistical error of the data points) is plotted on the right axis. To assure comparability of the size distributions their averaging times were chosen to be unambiguous with respect to LWC for in-cloud (mean LWC= 0.089 gm^{-3}) and out-of-cloud conditions (mean LWC= 0.016 gm^{-3}) within a short time interval (in-cloud: 19 September 2012 15:26–16:25 UTC; out-of-cloud: 19 September 2012 17:30–18:00 UTC). The vertical lines indicate the diameters of zero activation (D_0), 50% activation (D_{50}), Hoppel minimum (D_H), and full activation (D_f).

Table C1: Different combinations of hygroscopicity parameters (κ_a, κ_{cut}, κ_{mean}, κ_{AMS}) and activation threshold diameters (D_f, D_{50}, D_H, D_0) used to determine the cloud peak supersaturations reported in Table 2 by Köhler theory calculations as outlined in Appendix A and B.

	D_f = 191.1nm	D_{50} 58.6nm	D_H = 60.4 nm	D_0 = 37.7 nm
κ_a	0.51	0.19	0.19	0.19
κ_{cut}	0.48	0.20	0.20	0.20
κ_{mean}	0.3	0.3	0.3	0.3
κ_{AMS}	0.45	0.45	0.45	0.45

Table C2: Lower bounds, average values and upper bounds of cloud peak supersaturation (S_{low}, S_{avg}, S_{high}) obtained by Köhler theory calculations assuming different types of hygroscopicity parameter (κ_a, κ_{cut}, κ_{mean}, κ_{AMS}) as reported in Table 1 (SMPS method; Sect. 3.2). The values displayed in the second-last line represent the arithmetic mean ± standard derivation of the preceding four lines (SMPS method average). The supersaturation values displayed in the last line were obtained without assumptions about particle hygroscopicity (arithmetic mean ± standard derivation; CCNC method; Sect. 3.1).

	S_{low} (D = D_f, κ) [%]	S_{avg}(D =D_{50}, κ) [%]	S_{avg}(D =D_H, κ) [%]	S_{high}(D =D_0, κ) [%]
$S(D, \kappa = \kappa_a)$	0.07	0.68	0.65	1.31
$S(D, \kappa = \kappa_{cut})$	0.07	0.66	0.63	1.28
$S(D, \kappa = \kappa_{mean})$	0.09	0.54	0.52	1.05
$S(D, \kappa = \kappa_{AMS})$	0.08	0.44	0.42	0.86
S(SMPS)	0.08 ± 0.008	0.58 ± 0.10	0.56 ± 0.09	1.13 ± 0.18
S(CCNC)	0.19 ± 0.06	0.51 ± 0.06	0.51 ± 0.06	-