Atmospheric Chemistry and Physics Discussions

Interactive comment on "Size-dependent activation of aerosols into cloud droplets at a subarctic background site during the second Pallas Cloud Experiment (2nd PaCE): method development and data evaluation" by T. Anttila et al.

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Interactive Discussion

Discussion Paper

The study addresses the prediction of cloud condensation nucleus (CCN) number concentrations. Both a theoretical approach is introduced and further more this is applied to a data set that has been acquired during the 2nd Pallas Cloud Experiment. The authors present a new methodology to calculate the number of activated cloud droplets for a given size distribution. They perform numerous sensitivity studies in terms of chemical composition (hygroscopicity), size-resolved composition, and mixing state in

order to analyze the importance of these particles to CCN number prediction.

Unlike previous CCN studies, the present study aims to present an 'overall' approach that can be applied to the chosen and similar data sets without taking into account detailed knowledge on composition etc. At this point of research, it is certainly of high interest to do analyses in order to identify the key factors that affect CCN numbers. Thus, the topic certainly fits into the scope of ACP. However I suggest to revise several sections before publication.

General comments

The authors should make clear throughout the manuscript that they describe CCN prediction and not cloud drop number prediction. They mention that kinetic limitations on drop growth are not taken into account (p. 14529, 1st paragraph), however, later on they talk about 'a parameterization of cloud formation' (p. 14536, Section 4.4.). It has been discussed in several prior studies that in a cloud the supersaturation is controlled by updraft velocities (and other dynamic processes) and the number of cloud droplets is usually smaller than predicted based on equilibrium studies (i.e. solely assuming Koehler theory). The authors discuss briefly effects of entrainment and how those could be incorporated into their approach. However, they state that for the discussion of data 'no entrainment took place'. In addition, it seems that their consideration of entrainment only holds for inhomogeneous mixing. It has been discussed in the literature that homogeneous mixing processes might be as common as inhomogeneous mixing [Chosson et al., 2007]. How 'valid' is the application of the parameter x in Section 1.3.2 if homogeneous mixing or a combination of both processes occur? Regarding the facts that (i) the entrainment effects are not discussed by using the measured data and (ii) the effects of updraft on supersaturation and reduction of activated particles (cloud drops) is not taken into account, I suggest that the authors state clearly that their approach is only new in terms of CCN predictions and the effects of dynamics (entrainment, updraft) are not explored.

Specific comments

p. 14520, l. 18/19: Do the authors define 'hygroscopicity' as the quantity that is referred to as 'kappa' [Petters and Kreidenweis, 2006] or 'B' (= numerator in Raoul term of Koehler equation)? At least they should define it later in the manuscript in order to make clear how the calculate specific values as hygroscopicity.

p. 14521, l. 26 ff: As in my general comments, the maximum supersaturation in a cloud is (mainly) controlled by dynamics. Thus, the number of activated particles in an HTDMA does not necessarily give information on the 'real' supersaturation in a cloud since due to limited growth time, less particles will be activated.

p. 14524, Eq. 1: As it is written here, Eq.-1 describes the fraction of particles within a size class i that is activated at Smax. I think that Eq.-1 should be written in a form that expresses the total number concentration as a fraction of the total size distribution and not only as the fraction within one size class. Only then, the following sentence the maximum supersaturation in a cloud can be estimated (i.e. the condensation term if the dynamics term, ie. updraft would be known).

p. 14524, l. 14: The conversion from volume into mass fraction implies a given solute density. In addition, you assume ammonium sulfate as a solute. Is the analysis sensitive to this assumption at all?

p. 14527, l. 4/5: The reference to [Shulman et al., 1996] should be added here.

p. 14527, Eq.-8: Is there a physical explanation for the exponent -1/2 of the partitioning factor f?

p. 14529, l. 5: I agree that reliable information on updraft velocities is sparse. The text reads as if it is harder to obtain data on updraft than on entrainment from ground base measurements. Is this true? In any case, as suggested above, I think that sections 1.3.2 and 1.3.3. only distract form the main focus, namely CCN prediction, of the paper and should be only shortly mentioned if not removed at all.

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p. 14532, l. 4: Has the smaller growth factor for marine particles been observed in prior studies? As a 'first guess' I would expect that due to their high fractions of NaCl their hygroscopicity should be highest. Or might a small growth factor by indicative of organic material?

p. 14534, l. 9: How exactly can growth factors be measured? In other words, can a difference of 0.01 ion calculated growth factors be observed in measured growth factors as well?

p. 14536, l. 11ff: Again, the 'thought experiment' of changes in particle water uptake as a response to a change in Smax is only useful for equilibrium conditions since in a cloud a change in the loss term (i.e. particle's water uptake) will affect supersaturation. This point should be iterated here.

p. 14537, l. 8/9: Figs. 4-6 show nicely how differently hygroscopic particles will affect the activated fractions within the respective size classes. I wonder whether it might be possible to incorporate the number of particles in each size class somehow to the plots. This could be done e.g. by adding a second x-axis to the plots containing the number concentrations. Such a plot would highlight the importance of small variations in hygroscopicity at different parts of the size distribution: Whereas at large size, the number concentration is small, and, thus, it does not make a huge difference for the total activated CCN number whether 80% or 100% are activated, such a difference in activated fraction around 100 nm might affect the CCN number significantly. I realize that all this information is somehow included in Figure 1 and Table 4, but an overall figure would be nice.

p. 14539, l. 5/6: How realistic is the extrapolation of growth factors to larger sizes? At small sizes, growth factors (of particles of the same composition) might be biased to smaller sizes due to the Kelvin effect. However, above ˜ 100 nm this effect should be negligible. Thus, how would the results be affected if a horizontal line for the growth factors above ˜ 100nm would be assumed?

Table 4: Could you add the soluble fractions to the table that correspond to the various growth factors?

Technical comments

p. 14520, l. 21: Either '...activation profiles carry' or '...activation profile carries'

p. 14525, l. 19: Section 2.2. does not exist. Check also the remainder of the manuscript as in several places the numbering seems wrong.

p. 14527, l. 25: surface-active (not 'activate')

Appendix: Add 'GSD' to the list

Tables 1 and 3: Check the symbols: Use either A(ait) or N(ait) consistently.

Figure 2: Replace 'hollow symbols' by 'open symbols'

Additional references

Chosson, F., J.-L. Brenguier, and L. Schüller (2007), Entrainment mixing and radiative transfer simulation in boundary layer clouds, J. Atmos. Sci., 64, 2670-2682.

Petters, M. D., and S. M. Kreidenweis (2006), A single parameter representation of hygroscopic growth and cloud condensation nuclei, Atmos. Chem . Phys.

Shulman, M. L., M. C. Jacobson, R. J. Charlson, R. E. Synovec, and T. E. Young (1996), Dissolution behaviour and surface tension effects of organic compounds in nucleating droplets, Geophys. Res. Lett., 23, 277-280.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 14519, 2008.