Atmospheric Chemistry and Physics Discussions

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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.

T. Anttila et al.

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We would like to thank the reviewer for his/her constructive comments. Detailed replies to all remarks are given below.

General comments

R: 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

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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.

We acknowledge the problems that were mentioned by the referee, especially those related to entrainment. According to the suggestions, we have substantially revised the corresponding sections and merged them together. In the revised version, we have omitted the previous approach and discuss on the effects of entrainment briefly as follows:

"There is no straightforward way to extend the method to account for entrainment in a quantitative manner. In ground-based studies, however, possible errors caused by a dilution of the measured air masses can be avoided by checking that the activated fraction of particles approaches unity at a diameter range 300 nm. This is because entrainment leads to a dilution of cloud air and hence to a partial activation of larger particles (Noone et al., 1992)."

Specific comments

R: 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.

The term 'hygroscopicity" is used, in general, to refer to the tendency of particles to take up water. It can be quantified e.g. as a hygroscopic growth factor at a specific water activity or as a water soluble fraction of the particle dry mass. This is now clarified in the revised version of the manuscript.

R: 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.

According to this comment, we have re-phrased the second part of the paragraph as follows:

"Furthermore, on the basis of activation and H-TDMA measurements the fraction of particles capable of acting as CCN can be determined as a function of the water vapour supersaturation by applying Köhler theory. A comparison of the predicted size-dependent activation curves with the measurement results has been shown to give insight into the peak supersaturation reached during the cloud formation, on the adiabacity of the sampled cloud air and also on the possible presence of organic films on the particles (Svennigsson et al., 1994, 1997; Mertes et al., 2005)."

R: 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).

In order to motivate the considerations preceding the presentation of eqs. 6 and 7, we have revised the manuscript by rephrasing the sentence following eq. 1 as follows:

"We first derive an expression for the fraction of activated particles in the size bin i, AF_i^*, and then present how the maximum supersaturation reached during the formation of observed cloud(s), s_{max}, can be estimated by calculating activated fractions AF_i^* in each size bin i."

R: 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?

This was a typo, the correct expression is "water-soluble volume fraction". This has been corrected. Also, the results are not sensitive to the choice of the model salt provided that its properties are used consistently in calculations. We mention this before introducing eq. 2 in the revised version of the manuscript.

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

This has been done.

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

The exponent -1/2 arises because the critical supersaturation is inversely proportional to the square of the particle soluble mass.

R: 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.

The discussion has now been shortened, and the sections 1.3.2 and 1.3.3 have been

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merged in the revised version.

R: 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?

We have now omitted the presentation of the general features of the experiment campaign, as suggested by the other referee, and consider only the individual cloud cases. However, the preliminary results seem to suggest that 1) particles in marine air masses contained significant amount of organics and 2) the particle organic content is correlated with low hygrocopic growth factors. Here we do not wish to speculate about possible explanations, but this topic will be pursued further in a separate analysis.

R: 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?

The measurements errors are typically 1%-unit for the water vapor saturation ratio and 2% for growth factors (Joutsensaari et al., 2001). The error estimates are now mentioned in the manuscript in section 3 ("Experimental methods" section). Consequently, a difference as small as 0.01 cannot be probably discerned.

R: 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.

We have mentioned this point in the last paragraph of the section, so we feel that the issue has been adequately addressed.

R: 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

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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.

The figure 1, which showed the average size distributions for each air mass type, has been replaced by a figure showing the total and interstitial particle total and interstitial size distributions for each analyzed case (Figure 2 in the revised version). We have displayed the information in a separate figure since imposing it into figs. 4-6 would have made the figures hard to comprehend.

R: 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?

We have investigated this issue and added the following paragraph to the section 4.2.:

"The sensivity of the results to extrapolating the measurement data to the diameter range 150 nm was assessed by repeating the calculations with the mean HG factors and GSDs set equal those for 150 nm particles at the particle size range 150 nm. As a result, the estimated s_max and predicted number of activated particles change only up to 4 and 7%, respectively. Such a small sensitivity shows that the following results are not probably affected by the lack of measurement data for 150 nm particles."

R: Table 4: Could you add the soluble fractions to the table that correspond to the

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various growth factors?

First, here the referee meant probably Table 2, since Table 4 does not display any growth factors.

Since the soluble fractions were not used directly in calculations and because they depend on the chosen model salt (ammonium sulfate in this case), we believe that it is sufficient to give the range of soluble fractions that correspond to the range of HG factors. They span the value range 0.13 – 0.49. This is now mentioned in the manuscript when Table 2 is introduced.

Technical comments

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

R: 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.

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

R: Appendix: Add "GSD" to the list

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

R: Figure 2: Replace "hollow symbols" by "open symbols"

All these comments have been addressed in the revised version of the manuscript.

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

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