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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.

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 presented theoretical approach contains some assumptions, e.g., about the size dependent surface tension or influence of entrainment but they are clearly mentioned and mostly sufficiently discussed by the authors. Only concerning the non-

consideration of kinematic limitations (section 1.3.3) in the computational tool, a more quantitative statement about the possible consequences on the data evaluation would be helpful to gain more faith in the application of the presented approach.

We have added the following discussion regarding the potential impact of kinetic limitations: "...here it suffices to note that regarding clean continental and marine air masses that prevailed during the field campaign discussed in this study, errors in the predicted number of cloud droplets are expected to be 15% at maximum (Nenes et al., 2001). This error range is comparable to the experimental errors in inferred fractions of activated particles (see section 4)."

R: Rather instructive is section 4.4 where sensitivity studies concerning size dependent hygroscopicity, change in hygroscopicity and influence of mixing state are presented. What is missing here is an evaluation of the derived results with regard to the cited publications in this context, which should be included.

In section 4.4, we have included separate paragraphs following each sensitivity study, where our results are discussed in the context of the previous findings.

R: One drawback from the experimental point of view is that the reference of experiment overview and results is only a conference proceeding (Komppula et al., 2006). Especially the working principle and sampling efficiency of the total air inlet needs to be discussed in detail, since the activated number fraction is determined by the difference total minus interstitial and not directly by means of a ground-based counterflow virtual impactor (CVI) like in other studies of that kind. Due to the insufficient experiment description, it is not clear, whether cloud microphysical probes measuring liquid water content (LWC) and drop concentration were operated in this field experiment, which is actually mandatory for cloud measurements. In order to strongly justify the experimental concept drop concentration should be compared to the concentration of activated particles for the selected cloud events. Moreover the time course of the LWC during the selected cases provides indications whether the thermodynamic conditions

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during the cloud period remained rather constant. The possibility to revert to such useful supporting data must be addressed by the authors.

We now describe the inlet system in more detail in the "experimental methods" section as follows:

"Total air inlet has the same construction as used and designed by Weingartner et al. (1999). The inlet consists of heated inlet and a heated snow-hood. The inlet design allows cloud droplets having a diameter $<$40 μm to enter the sampling line. The heating dries all cloud droplets as soon as they enter the sampling system and both cloud residual and interstitial particles are measured. According to the calculations of Weingartner et al. (1999), the sampling efficiency of the total inlet system is well above 95 % in the size range 10-40 um which is relevant for aerosol/cloud interaction studies. No cloud microphysical probes such as a FSSP (Forward Scattering Spectrometer Probe) were operating during the considered time periods, and hence there is no direct information on the liquid water content (LWC) or on the cloud droplet effective radius. Such information would be useful in interpreting data, but not necessary, however, since the focus here is on the size-dependent activation of ambient aerosols."

R: Unfortunately, the model formulation (section 1.2) is rather incomprehensively presented. This comprises the insufficient introduction of equations and the use of uncommon key symbols. In detail, this will be addressed in the specific comments.

We have substantially revised this section, as detailed in our replies to the specific comments (please see below).

R: Another point is that the authors should more pronounce the intended utilization of their approach. Is it really limited to a post-analysis of ground-based cloud measurements inferring the peak supersaturation of different case study clouds? Or will their tool have further applications in cloud models to improve predictions of certain cloud parameters?

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Our approach is not limited to a single application; for example, it can be used to parameterize CCN spectra for use in large scale models and also in connection with CCN measurements where CCN spectra were measured as a function of the water vapour supersaturation. These features are now brought out after introducing equation 7 in section 1 (section 2 in revised version) and also in the "summary and conclusions" section.

Specific remarks

R: P. 14520, L. 2: It is more precise to speak of "aerosol particles" than of "aerosols". This should be changed throughout the text where appropriate.

Given the scope of the journal ACP and the common usage of language in the articles appearing in ACP, we believe that using the expression "aerosol(s)" or "particle(s)" is sufficient in this context.

R: P. 14520, L. 5 (abstract) and P. 14538, L. 26 (conclusion): Despite the addition in brackets “in terms of the water activity”, the authors should avoid the still misleading item “chemical composition”, since they did not present any chemical identification in this manuscript. Better use “particle soluble fraction” or similar.

We have now avoided the use of the term "chemical composition" by replacing it with "hygroscopicity" or "water soluble fraction" where appropriate.

R: P. 14523, L. 15-17: Since the use of cumulative HG factor distributions is pretty unusual their convenience for the actual study needs to be further explained.

We have now motivated the use of the cumulative distributions by stating that "As seen later, the activated fraction of particles can be treated also as a cumulative distribution, and this makes the use of cumulative HG distributions convenient."

R: P. 14523, L. 17: How are the distributions fitted for particle radii for which no hygroscopic growth is measured?

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This can be done in cases where there are hygroscopic growth measurements for several other diameters. The parameters that are obtained from the hygroscopic growth data, such as peak hygroscopic growth factors, can be then inter- or extrapolated to the needed particle diameters.

We now mention this point in the first paragraph of the section.

R: P. 14523, L. 14 – P. 14524, L. 4: Eq.1 has no direct relation to the Köhler theory, i.e. this statement needs to be reworded.

We have omitted the reference to Köhler theory.

R: P. 14524, L. 6-9: These statements sound trivial. If they are not they need more explanations. Should s (used two times here) be replaced by smax at least the second time.

Statement regarding the equality of AF and K is not necessarily true because of the kinetic limitations and because some large particles may reach cloud droplet sizes even though they have not activated according to Koehler theory (Nenes et al., 2001). Therefore this is not a trivial statement, and we also bring out this in the revised version.

Also, the symbol "s" should have been "s_max". This has been corrected.

R: P. 14524, L. 19: In connection with Eq.2 GRmeas is not a cumulative distribution of the HG factor but the growth factor itself, thus another key symbol, like the common gf for growth factor should be used.

According to this suggestion, we have replaced the symbol "GRmeas" with "gf".

R: P. 14524, L. 25, Eq.2: First, it is not so straight forward how Eq.2 is obtained from Eq.1 with respect to the relation of SF and GR. Second, what is the meaning of the squared brackets? Thus, the derivation of Eq.2 needs much more explanations. Third, what does that mean, the distribution of the particle water-soluble volume fraction SF as a function of the soluble volume fraction ε? Fourth, why does ε does

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not have an index i since it also changes with particle size? All these issues needs to be addressed.

First, we have now explained more in detail how eq. 3 is derived from eq. 2 (We presume that this was the intention of the referee, and not to explain how eq. 2 is derived from eq. 1 which have no connection). To be more specific, when introducing the parameter SF, we now bring out that it is a function of v so that SF_i(eps) is the fraction of particles in the size bin i that contain less water-soluble volume than eps. Similarly, when introducing the parameter GR we now mention that it is a function of gf so that GR_i(gf) is the fraction of particles in the size bin i that have a hygroscopic growth factor smaller than gf. After introducing (2), we note that (2) allows for converting hygroscopic growth factors to the corresponding soluble volume fractions. From this and from the definitions of GR and SF it is easy to see how (3) follows from (2).

Second, the brackets indicate that the argument of the function GR_i is here another function, gf_i. This is now explained in the manuscript.

Third, as described above, SF_i(eps) is the fraction of particles in the size bin i that contain less water-soluble volume than eps, and eps in equation (2) indicates the soluble volume fraction which corresponds to a hygroscopic growth factor gf. We have revised the manuscript accordingly to remove the ambiguity on the definition of SF.

Fourth, eps is a variable that appears in (2), and the soluble volume fractions are characterized here by size class-specific distributions SF_i(eps). This point is now clarified in the manuscript.

R: P. 14525, L. 12, Eq.5, upper part: Again the derivation or origin of the upper part of Eq.5 is unclear. It seems that the authors assume K(s) = 1 – SF(ε). This must be explained. Again the meaning of the squared bracket is not clear.

We have added the following explanation before introducing eq. 5:

"According to the definitions of SF_i and K_i, K_i(s) can be expressed in terms of

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SF_i(s) as follows: K_i(s)= 1-SF_i(eps*) where eps* is the particle soluble fraction needed for activation at a supersaturation s. Using eq. (3), this relation can be further expressed as: K_i(s)= -GR_i[gf_i(eps*)]. The relationship between eps* and s is, in turn, obtained from eq. (4)."

This, together with other revisions discussed above, should make the derivation transparent.

Second, the brackets indicate that the argument of the function GR_i is here another function, h_i. This is now explained in the manuscript.

R: P. 14530, L. 15 – 25: Since many readers will not be familiar with the surrounding of the measurement site, a map of the region including typical back-trajectories for the EUR, MARINE and MIX air masses would substantially improve the comprehensibility of this text passage. From the text description it is not clear whether back- trajectories of the same air mass category always came from the same directions or could arrive from completely different regions. This needs to be additionally addressed.

We have now added a figure which shows the location of the measurement site, its surroundings as well as trajectories illustrating each air mass type. The figure shows that marine air masses originate typically from west or north while European air masses originate from south or east.

R: P. 14531 and 14532, section 3: It is not clear why the authors included this chapter into the manuscript, since aerosol parameters of the case studies examined later on are more or less far away from the general characteristics presented here when comparing Tabs.1 and 3. Instead or in addition of presenting the general air mass classified particle number size distributions in Fig.1 it would be much more meaningful to illustrate the number size distributions (total and interstitial) of the cloud events that were analyzed in the next section. So this section must be better motivated by the authors and the figures changed or extended.

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We have removed the general discussion on the results of the campaign as well as the table and figure illustrating these results (Table 1 and Figure 1). Instead, the figure 1 has been replaced by a figure illustrating the total and interstitial size distributions for each case.

R: P. 14532, L. 23ff: Since the presence of hydrophobic particles could be an expected reason for a partial activation of large particles it is unclear why the authors did not measure the HG of larger particles. This can be easily done up to at least 250 nm (e.g., Lehmann et al., Atmos. Environ., 39, 4257-4266, 2005). This should be commented.

Yes, this is possible with the setup used in the experiment. However, hygroscopic growth of larger particles was not measured due to technical problems associated with using larger voltages in the first DMA. This is now mentioned in the manuscript.

R: P. 14533, L. 22 – 23: Although not required for the presented approach, the reader is curious to get some idea about the chemical composition of the aerosol particles related to the different air masses, especially for the MARINE case. Is there any chemical information from 2nd PACE measurements or other studies from that area so that the authors could speculate how the HG factors could come about? Again that would be particularly interesting for the low HG factors for the marine air masses.

We have now omitted the presentation of the general features of the experiment campaign and focus only on the case studies. 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 study.

R: P. 14547, Table1: The column Aacc/Nact announced in the Table caption is missing. For completeness another column for Nacc should be added, whereas the column Vtot can be erased. The difference in particle volume is also clear when including Nacc. Coming back to a comment above the meaning of section 3 and thus the information

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given in Table1 for the conclusions in this paper is not clear. Thus, section 3 (including Table1) should be more clearly motivated or deleted.

Table 1 and section 3 have been removed.

R: P. 14548, Table2: There is a layout problem between the columns for 80 and 100 nm. Again there is the same motivation problem as already identified for Table1. Instead or in addition it would be worth to have such a Table for the 4 selected cloud events beside their graphical presentation in Fig.2.

This has been now replaced with a similar table showing the mean HG factors for the analyzed cases.

R: P. 145549, Table3: This Table should include much more parameters, like HGF for all sizes, Nacc, Nacc/Ntot, Aacc/Nact. Mandatory is the declaration of D50 for each case, which is not explicitly mentioned in the text or given in Fig.3. If available also measured cloud microphysical and meteorological parameters like LWC, drop concentration, temperature would be helpful. Also the computational results, like the predicted drop concentration and estimated maximum supersaturation should be included. Then of course the Table must be transposed.

The table has been revised include all the mentioned information, excluding LWC and droplet concentrations because no measurement data was available for these quantities during the analyzed cloud events due to the lack of FSSP measurements for that time period (please note that the number of this table is 1 in the revised version). Also, the mean HG factors for all sizes and for all cases are now shown in Table 2.

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