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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.

Anonymous Referee #2

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Review of the paper „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. Antilla et al.

The scientific content of the manuscript is a relevant topic within the scope of ACP and with some minor changes (requested in the general and specific comments below) it is appropriate and recommended for publication. However, it is absolutely essential that

section 1.2 where the general formalism of the model approach is introduced will be substantially revised.

In general, the manuscript is well written with respect to language and structure. Only the numbering in the text links needs to be checked. For example, section 1.3 seems to be referenced as section 2.2 in the text although section 2.2 does not exist at all.

General comments:

This paper presents a computational scheme that uses the measured size resolved hygroscopic growth of aerosol particles in order to predict the fraction of activated particles as a function of particle size. By comparing these predictions with the measured activated fractions inside ground-based clouds in northern Finland estimates for the peak supersaturation in these clouds and information of the aerosol mixing state are obtained.

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.

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.

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

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

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.

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?

Specific remarks

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.

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.

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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.

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

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.

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.

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.

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

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.

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-

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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.

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.

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.

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.

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

P. 14548, Table2: There is a layout problem between the columns for 80 and 100 nm.

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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

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.

Technical corrections:

This is already included in the “specific remarks”.

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