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Geoscientific
Model Development
Discussions



# *Interactive comment on* "Meteorological and trace gas factors affecting the number concentration of atmospheric Aitken ($D_p$=50 nm) particles in the continental boundary layer: parameterization using a multivariate mixed effects model" *by* S. Mikkonen et al.

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We thank the reviewer for the comments and suggestions. Our point-by-point reply to these comments is below.

Note to editor: While this issue was not pointed out by the reviewers, in the original text we recognized that the term "model" was being using used both to describe the statistical model used to develop our parameterization as well as the chemical transport

model (GLOMAP) into which the parameterization is incorporated. To eliminate potential confusion, we modified the text so that “statistical model” is used when discussing the development of our parameterization, and “model” is used to describe GLOMAP or other models that will incorporate the parameterization.

Anonymous Referee #1

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This paper applies multivariate data analysis in order to derive a simple parameterization for atmospheric cloud condensation production associated with atmospheric aerosol formation or primary emissions of very small particles. The parameterization is evaluated over central Europe and compared with large-scale model prediction. The paper is definitely original, and the developed tool might be useful for the scientific community. The text itself is clearly written and well organized. A few issues should be addressed before the paper can be accepted for publication in GMD.

Detailed comments:

Page 1188, lines 18-25. As a rule of thumb, the size limits of 50 and 100 nm may be good estimates for the minimum particle diameter causing the indirect and direct radiative effects, respectively. However, it is well know the effective CCN activation diameter depends on particle size and composition, along with the selected value of supersaturation. As a result, the minimum CCN activation diameter probably ranges between about 50 and 100 nm for most boundary layer clouds. I am not against the selection of 50 nm here, but the authors should explicitly bring up this variability in the text with appropriate literature references. Similarly, it should be pointed out that direct radiative effect start to become important after 100 nm (there is no sharp size cut here). Furthermore, it should be stated whether the authors refer to particle dry or wet diameter, since the latter one is sensitive to the local relative humidity.

- Appropriate references will be added to text



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- Both reviewers encountered some difficulties in understanding our motivation for choosing N50 for this study, therefore we will provide an explanation here and will revise the text on page 4, lines 5-13, as well. We chose to study N50 since this is the minimum size for which any impacts of aerosols on climate are expected (as the reviewer states, only indirect impacts may be pertinent at this diameter). Our motivation for calculating N50 is that models that utilize this parameterization can initiate their aerosol modules at this diameter, thus saving computational resources over models that initiate aerosol formation at smaller diameters. We do not suggest that this population of N50 is involved in CCN activation: that depends on factors such as temperature and water vapour supersaturation as well as aerosol physico-chemical properties. These factors, combined with aerosol dynamical processes such as coagulation and evaporation/condensation, ultimately determine the climatic impact of N50 and are expected to be calculated in the model. Thus, by incorporating our parameterization we provide modellers an opportunity to lower the computational cost of their calculation without sacrificing accuracy; however, the environmental conditions and aerosol chemical properties from the model will ultimately determine the direct and indirect impacts of these particles.

- As page 5, line 18 states, we have parameterized this model using the dry diameter.

Besides atmospheric measurements and model investigations, theoretical frameworks have been derived to investigate the efficacy by which nucleated particle produce CCN in the atmosphere (e.g. Pierce and Adams (2007) Atmos. Chem. Phys. 7, 1367-1379; Kuang et al. (2008) GRL 110, doi:10.1029/2009GL037584, and references therein). This should be briefly mentioned in the manuscript.

- Suggested references have been added to the introduction.

The scientific/technical objectives of this paper should be explicitly stated in the Introduction. One of the aims have been mentioned on page 1192 (lines 13-15), but that is too late.



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- The objectives are now pointed out in the introduction with text: "The main objectives of the study are to find the factors affecting the growth to, and primary production of, particles that can be considered the minimum potential CCN size and to find a parameterization which can be used as a part of a larger atmospheric model to predict the concentration of climatically active particles."

I understand that the analysis performed in section 3.4 was meant as a preliminary test of the performance of the new tool/parameterization in a large-scale modeling framework. Therefore, only a time period of one month and three stations were used for the comparison. I see a potential problem here: since the comparison was made against data from the same locations based on which the parameterization was developed, isn't there a danger of getting a biased (too positive) view on the performance of the parameterization? Could it be possible to run the model for a month in some later year when, for example, more size distribution data associated with EUSAAR or EUCAARI measurements are available? Is it really too expensive to run a model for a few more months, so that the comparison to the measurement data would be easier (there are many more 1-year long data sets).

- We have now included a comparison of the original GLOMAP model run to 15 EUCAARI sites. Note that the EUCAARI measurements are from years 2008 and 2009, while the original model simulation is for year 2000. However, rerunning the model for a later time period would be quite laborious and time consuming, since it would require rerunning also the coupled chemistry aerosol model to obtain new offline fields for the input oxidants. We have therefore not made any new simulations for the revised manuscript but explain the uncertainties arising from the use of different model year more carefully in the text.

- Accordingly, we have changed in the text on page 15, line 24:

"Figure 7 compares the model-predicted potential CCN concentration (Dp > 50 nm) against measured April mean data at Melpitz, SPC, Hohenpeissenberg, and 15 other



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sites. For the three sites analysed in this study, we present averages of several years of measurement data (i.e., multi-annual averages), since the analysed measurement periods at the three sites did not overlap (see measurement periods in section 2.1). The data for the other 15 sites is from the European Integrated project on Aerosol Cloud Climate and Air Quality Interactions (EUCAARI) during April 2008 and/or 2009, and were chosen because of their comprehensiveness. Note that since the model was run only for April 2000 (due to computational expense of a global model) one can expect only a rough agreement between the model and measurements. However, we wanted to include the EUCAARI data in order to demonstrate that the statistical parameterization gives reasonable results also outside the geographical domain for which it was derived."

"Of the 15 EUCAARI sites, using the parameterisation instead of the baseline model set-up brings the predicted CCN much closer to observations at 7 locations and deteriorates the agreement clearly at 4 locations. At 3 sites (K-Puzsta, Kosetice and Finokalia) there is relatively small difference between the baseline and the parameterized runs. At Waldhof the observed CCN is approximately halfway between the predicted values from the baseline and parameterized runs.

While this preliminary test of the parameterisation against observations is incomplete in that it does not simulate the exact years of the observations, it does indicate that the derived parameterisation has potential to describe CCN formation at very different environments from Arctic to polluted rural. These results give confidence to apply the statistical framework also to measurements from other sites in order to further improve the derived parameterisation."

Finally, why to compare only at 50 nm? Both primary and secondary particles grow beyond this size, so additional information on the performance of this parameterization would be obtained if also other sizes were compared (e.g. 80 and 100 nm).

- Note that we present "potential CCN" (Dp> 50 nm), i.e. the concentration of all parti-



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cles larger than 50 nm, and not particle size distribution function at 50 nm. This lower limit for CCN was chosen since it gives more weight to the actual parameterization than higher cut-offs. This is because at larger sizes the role of primary emissions to the high end of Aitken/low end of accumulation mode becomes more and more important with respect to the particles grown from below 50 nm (which are the only ones represented in the parameterization). While possible shortcomings in the aerosol primary emission inventories used in the model for particles over 50 nm will bias the comparison also for potential CCN (> 50 nm), this is even more so the case for higher cut-offs.

- "CCN" will be changed to "potential CCN" where appropriate

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