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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.

Anonymous Referee #2

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Interactive Discussion

Discussion Paper

General Comments:

A statistical parameterization to predict the number density of 50 nm particles over Europe has been developed as a function of time, relative humidity, SO2, NO2, O3, temperature, condensation sink and non-new particle formation days. The rationale is that 50 nm particles can act as cloud condensation nuclei (CCN), and that detailed microphysical models to predict their concentration are computationally intensive relative to this parameterization. The paper addresses a modeling problem appropriate for GMDD.

While an interesting treatment, the main problem I had with the paper is the assumption that since 50 nm particles can act as CCN, they do act as CCN. Clearly this is not always the case, depending on meteorological conditions, pre-existing aerosol number concentration, and hygroscopicity of the aerosol (which the authors admit is highly variable at 50 nm). With >50 nm aerosol concentrations as high as 3000 cm-3 at the polluted sites in their data set, it is unlikely that 50 nm particles will activate under many of these conditions. In fact, they are likely to be more important as CCN in clean sites. While I am sure the authors recognize this, the terminology used in the paper slips from specifics of the 50 nm measurements to “CCN concentration” in the Results section. I would suggest calling these “potential CCN” in this section.

- This comment is similar to that of the first reviewer, and our response to that comment is applicable here. In summary, we have revised the text on page 4, lines 5-13 as follows:

“A prime motivation for the current study was to derive a statistical relationship between meteorological, trace gas, and aerosol properties and 50 nm particle concentrations. The resulting parameterisation could then be used in large scale models so that no computing resources go into aerosol dynamics below 50 nm. We do not suggest that this population of 50 nm particles are themselves climatically important: this depends on factors such as temperature and water vapour supersaturation as well as aerosol

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physico-chemical properties. These factors, combined with aerosol dynamical processes such as coagulation and evaporation/condensation, ultimately determine the climatic impact of this population and are expected to be calculated in the model. Thus, by incorporating our parameterization we provide modelers 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."

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

Additionally, some estimate of the expected supersaturations reached in low level clouds in the regions of study should be made. How these compare with the critical supersaturations of 50 nm particles at the different hygroscopicities measured would aid in understanding how relevant the parameterization for 50 nm particles really is for cloud formation.

- This comment is probably based on the mistaken impression that our calculation of N50 infers that all of these particles act as CCN. We admit that this was not clear in the manuscript, however our response to the previous comment, we feel, will address this issue and probably make the current comment irrelevant (since the model that will use this parameterization should take into account the actual climatic relevance of these particles).

I agree with Referee 1 that parameterizations for other (larger) particle sizes would be of interest as well.

- Some tests with 100 nm particles have been made and the best predicting variables seem to be almost the same as for 50 nm particles but those results will not be presented here.

The rest of the paper is well written, but there are a number of minor points where terminology or details are not well defined, as listed below. Specific Comments:

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p 1188, line 1: “these new particles” are invoked without defining them earlier in the paragraph; earlier the discussion is on atmospheric aerosols in general rather than newly formed particles. p. 1191, line12: NPF days are listed here, but the definitions of event and non-event days are not given until the next section (2.2). Recommend rearranging things here for clarity, defining the terminology first.

- Sentence in p1188 has been changed into form "Theoretical frameworks have been derived to investigate the efficacy by which nucleated particle produce CCN in the atmosphere (e.g. Pierce and Adams, 2007 and Kuang et al., 2008) and recent atmospheric measurements and modelling studies have shown that new formed particles can affect concentrations of CCN (Ghan et al., 2001; Lihavainen et al., 2003; Kerminen et al., 2005). "

p. 1193; line 1: insert (CS)after “condensation sink”.

- Inserted.

p. 1194, line 9: What is the bin size used for N50?

- N50 is defined in section 2.1, and is actually and thus independent of bin size.

p. 1196, Eqn 3:Please clarify utime. Does this refer to time of day? Because based on Fig. 4, time of day is an important variable where the model does not seem to reproduce the variation very well. And a related question, was some measure of solar radiation specifically used in the parameterization?

- Here utime refers only to month and for clarification it will be renamed to um. Using different u-values for time of day (uh) did not give significantly better results in comparison to additional computational cost.

- Solar radiation was not directly used in the final parameterization but global radiation was used in predicting NPF days with discriminant analysis like stated in section 2.3. Global radiation was tested as a predictor in the model but the effect found out to be not significant.

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p. 1198, line 1-2: I don't understand how SO2 can be positively correlated "at all sites", but negative at SPC and Melpitz.

- The sentence is clarified into form "SO2 concentration had a significant positive regression effect in the summer at all sites but the effect was negative or zero in wintertime, especially in SPC and Melpitz."

p. 1198, 2nd paragraph: Wouldn't NO2 also be a general indicator of more anthropogenic pollution, and therefore correlate with higher N50?

- That is one possible explanation but as stated in the text we do not know what is the real reason for significance of NO2 in the model

p. 1200, line 15; p. 1203, line 25: The authors should define what they mean by "adequate". Adequate for what?

- The full sentence in question states: "An advanced statistical model structure was introduced and found to be an adequate tool to analyse tropospheric Aitken particle (Dp = 50 nm) concentrations and for making predictions based on in-situ meteorological (temperature, RH) and gas phase parameters (SO2, NO2, O3)." We feel that the answer to the reviewer's question lies in this sentence: our analysis shows that the model is adequate for representing 50 nm diameter particle concentrations in models.

p. 1211, Table 3: I am not sure what "multiannual averages" means. Are they several years of measurements averaged for April, in comparison to the model output for April 2000? As the authors admit, the time periods used in the table do not seem to be the same time periods as for the simulation, so can they really say that "the parameterization (test run) significantly improves the agreement with observations at Melpitz and Hohenpeissenberg"? Is there any way of looking at how well variation within certain time periods are duplicated, rather than just using a bulk median comparison?

- As the reviewer suspects, "multiannual average" refers to several years of measurements averaged for April. This is now clarified in the manuscript. Since the measure-

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ment periods for the available data did not overlap, we chose to use this average value instead of running a 2 month spin-up + April for three separate years (both for the aerosol and the chemistry model), which would be computationally quite expensive. For the same reason, running the model for several years at each site (which would be required to study the variation) is unreasonable for this type of a preliminary test.

- However, it is a fair concern that since we are not comparing exactly the same time periods in the model and the measurements, the formulation of the indicated sentence may be misleading. We have therefore modified it to:

“The parameterization (test run) brings the model results significantly closer to observations at Melpitz and Hohenpeissenberg: The baseline run predicts 110% and 52% higher than observed potential CCN concentration, respectively.”

- Furthermore, we have added a paragraph highlighting this issue: “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.”

Interactive comment on Geosci. Model Dev. Discuss., 3, 1185, 2010.

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