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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of the accuracy of analysis tools for atmospheric new particle formation" *by* H. Korhonen et al.

H. Korhonen et al.

hannele.korhonen@alumni.helsinki.fi

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We thank the reviewer for his/her comments and suggestions. Our point-by-point reply to these comments can be found below. The page numbers refer to the version published in ACPD.

"Maybe some of the defined symbols such as the different Δt could be introduced using a contour plot of a new particle formation event. This would be much more illustrative and improve the clarity."

We have added a new figure showing a contour plot of one simulated new particle formation event together with the corresponding nucleation and new particle formation rates and an illustration of the fitting procedure for Δt_{N3-6}.

In addition, we have added the following text to the end of the model description (section 2.1):

"Figure 1a shows an example of a DMPS-gridded distribution from one model run. It is worth noting that while the simulated event resembles measured atmospheric events closely in most respects, the modelled data is much smoother and lacks noise that is present in typical atmospheric data due to instrumentation and inhomogeneities in the measured air mass. The smoothness of the modelled data is evident also in Figure 1b which presents the simulated nucleation and particle formation rates together with the scaled concentration of 3-6 nm particles (N_{3-6}). Note that while the modelled N_{3-6} is used as an input in the analysis described below, the simulated $J_{1.5}$ and J_3 are used only for comparison with the respective predicted values."

And to step 1 of the description of the analysis tools (section 2.2):

"The fitting procedure is illustrated in Figure 1c, which depicts the simulated H_2SO_4 (blue line) and N_{3-6} (red line) concentrations. In this specific case, when the H_2SO_4 curve is delayed by 60 minutes and raised to the power 2.31 (black dashed line), it is evident that it correlates very closely with the simulated N_{3-6}."

"In general, I was missing some relation to measurements, e.g. comparison of uncertainties and so on. The main goal of these analysis tools is the analysis of measurements, thus also this evaluation should be connected to measurements."

We fully agree that the fundamental aim of the analysis tools is to get information about atmospheric nucleation from ambient measurements. However, connecting the obtained results to actual atmospheric events in a quantitative way would require us to know which of the 1239 simulated events, which differ from each other notably regarding the nucleation mechanism and vapour properties, resemble closest the atmospheric ones. Due to the lack of theoretical understanding surrounding atmospheric nucleation events, this is not currently possible.

To explain this, we have added to the conclusions:

"Unfortunately, quantifying the error that the analysis tools have caused in previous analyses of atmospheric data is not straightforward since we do not know which of the simulated events resemble closest the atmospheric ones. Since the tools perform very well for some individual simulated events and quire poorly for others, it is equally possible that the tools have introduced only minor error in atmospheric analyses or alternatively that they have misdirected our theoretical understanding regarding e.g. the nucleation mechanism. Currently, we cannot know if either is the case; however, our study raises the point that large errors are possible and thus caution should be practiced when interpreting the atmospheric data. "

"From the title I expected that different models developed during the last years were compared here. But as far as I understood, the paper is based on one model varying the different parameters only. From the title this is a bit disappointing and it promises too much. If there are really different tools included it needs to be explained. However, it would be a really good idea to estimate the effect of different models on the analysis of nucleation events. Do other groups use similar algorithms or are they completely different? It is probably not possible to include other models into this study within a reasonable time but a comment about the expected variation between different models would be nice. Do they all use similar mechanisms? This could be mentioned here. And a real comparison of different model could be the topic of another study."

The term "analysis tools" in the title refers to the earlier developed mathematical equations (in the manuscript: equations 6-11 and 14-16) that are being evaluated in this study, not to aerosol models. Since there are several tools, i.e. separate ones for calculating J_3, $J_{1.5}$ and growth rate plus in some cases several formulations, the plural form in the title is justified.

It is these tools alone that are used to analyse the nucleation events; the model is used

only to produce the event data for the analysis. Prescribed mathematical nucleation events are used since the nucleation mechanism and the growth of these events is known exactly; this is not the case with real atmospheric data. Given the aim of the study, a comparison to other models would not make much sense: when two well-coded aerosol models are given exactly the same prescribed input and equations to solve, they naturally give the same output.

"Here, the model was applied to modeled NPF events, i.e. the nucleation and growth processes were triggered by those processes included into the model. What's about real cases in the atmosphere? There is usually a variety of precursor gases available and I personally think that in many cases we have no clue what's going on there. Is there a parametrization covering these uncertainties in the model?"

We agree with the reviewer that the theoretical understanding of actual atmospheric nucleation events is poor. Because of this, our model runs cover 1239 different conditions with varying nucleation mechanisms, properties and concentrations of nucleating and condensing vapours and pre-existing particle populations (see Table 1 in the manuscript for details). Some of these conditions are bound to be closer to real events than others but our limited understanding of nucleation prevents us from telling which ones are which. However, this is not even crucial for evaluating the performance of the tools since the tools themselves do not make any assumptions about the nucleation mechanism or the vapour properties; thus they should perform as well for all (real or made-up) events regardless of the specific mechanism or vapours.

To explicitly acknowledge the possibility that there probably is a larger variety of condensing compounds in the atmosphere than in our study, we have added to the end of the first paragraph on page 26294:

"Note also that while we simulate only sulphuric acid and one condensing organic compound, in the atmosphere there may be several others (e.g., amines, several organic

compounds with different properties) contributing to the early stages of cluster growth (e.g., Smith et al., 2010). Their combined effect could cause even a stronger deviation from the constant growth rate assumption than simulated in this study."

"Page26296, line 23ff. The authors say that N3−6 is directly obtained from measurements in contrast to J1.5 which was calculated using several assumptions. Is this statement related to this study only or is it generally true? I understand that for simulated new particle formation events N3−6 should fit nicely to the predicted values, but is this true for measured cases?"

This statement refers to all measurements in which the lower detection limit of the size distribution instrument is at 3 nm (if the limit is higher, then the directly obtained number concentration is of some higher size range; e.g. N_{10-15} for cut-off diameter 10 nm). There are currently no instruments that can directly measure the nucleation rate in atmospheric conditions at 1.5 nm, and thus $J_{1.5}$ is always calculated using some assumptions.

N_{3-6} is not a predicted value either in the case of measurements or the model. Instead, it is the primary input taken from the data for the analysis tools. In the case of measurements, N_{3-6} is the measured number concentration in the 3-6 nm diameter range. As with most instruments measuring close to their cut-off region, it is possible and even likely that there are uncertainties associated with the data from the lowest channels of e.g. the DMPS instrument. However, this is a separate question from the performance of the analysis tools. In the very last paragraph of the manuscript, we mention the noise from the instruments as a possible further source of uncertainty in the analysis of actual atmospheric data.

"Finally, what do we learn from this study? How large are the uncertainties compared to other errors, such as those from measurements? Can you give a list of recommendations which settings should be used preferably in such a model? Some recommendations are between the lines, but it would be nice to put them

together."

We have tried to articulate the main message of the study more clearly by adding to the end of the abstract:

"Overall, while the analysis tools for new particle formation are useful for getting order-of-magnitude estimates of parameters related to atmospheric nucleation, one should be very cautious in interpreting the results. It is, for example, possible that the tools may have misdirected our theoretical understanding of the nucleation mechanism."

And to the conclusions:

"Overall, we conclude that the analysis tools have built-in assumptions which can cause uncertainties in the event analysis. While this uncertainty is in most cases within an acceptable order-of-magnitude limit, it is important to be careful when interpreting the data and drawing conclusions about e.g., nucleation mechanisms or temperature dependence of nucleation prefactors, etc. Unfortunately, quantifying the error that the analysis tools have caused in previous analyses of atmospheric data is not straightforward since we do not know which of the simulated events resemble closest the atmospheric ones. Since the tools perform very well for some individual simulated events and quire poorly for others, it is equally possible that the tools have introduced only minor error in atmospheric analyses or alternatively that they have misdirected our theoretical understanding regarding e.g. the nucleation mechanism. Currently, we cannot know if either is the case; however, our study raises the point that large errors are possible and thus caution should be practiced when interpreting the atmospheric data. "

As discussed in this added text, it is impossible to say at this point how large the uncertainties caused by the analysis tools are compared to those from the measurements.

For the same reason it is not possible to give a conclusive list of the optimal setting for the analysis tools. The recommendations that we can be certain of irrespective of the

actual atmospheric event type (i.e. event not suitable for analysis if nucleation mode contributes significantly to the coagulation sink; time delay between J_3 and H_2SO_4 should not be used; J_3 is most accurately captured with the Vuollekoski formulation) are already explicitly stated in the conclusions.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 26279, 2010.