

Interactive comment on “A numerical comparison of different methods for determining the particle formation rate” by H. Vuollekoski et al.

H. Vuollekoski et al.

henri.vuollekoski@helsinki.fi

Received and published: 17 June 2011

Major comments

In the introduction part, the author state that using a simplified model has advantages compared to analysis of measured data. This is due to the fact that measurement data is associated with numerous processes that interfere with the analysis, such as changes in air masses and boundary layer height. Then however, the authors still conclude that the derived results are applicable to analysis of measurement data. Can the author expand on this? How would the knowledge gained through this study be transferred into more complex systems where the omitted processes de facto are active? For example, can this statement be justified when the authors adopt highly

C15210

simplified assumptions regarding the nature of the organic compounds participating in the initial steps of the growth (i.e. I assume it is single low volatile compound)? The nature of this compound (thermodynamic and molecular properties) is furthermore not discussed in the MS. The reason why we make this statement of “simplified model has advantages compared...” is just because we know ‘the answer’ to our problem. We are aiming at developing/testing methods for estimating particle formation rates from measured size distribution evolution only. What would be a better way of testing these methods than a numerically generated event for which we know the answer??? The reviewer is correct in that some potentially significant processes are omitted. In any case, before now, we have not known how good the (already much used) methods are – even for these simplified dynamics. The reviewer is also correct in that the simplified treatment in our model for condensational growth may be very different from what happens in the atmosphere, including several vapors, each with different equilibrium vapor pressures etc. However, this does not affect our analysis much. Our aim is to test/develop formation rate estimation methods that rely on analysing the number distribution only (and not knowing anything about the growth-participating vapors etc.), especially to find out what are the possible causes of error in the already widely used equation 5.

As a main conclusion, the authors state that the often used power law dependence is inaccurate. As the MS is written, I do not see any clear support for this statement. The power law relation is used to connect the concentration of nucleating material ($[H_2SO_4]$) to apparent formation rate. The author’s base this conclusion on the fact that maximum in sulfuric acid concentration not necessarily coincide with maximum in J3. This statement is rather strong, and requires more attention in the MS. The authors should provide tentative explanations for this mismatch. This statement should be backed up by analysis of when and why the power law fails, and when and why it is applicable. What is the role of the initial cluster size? What is the role of the nature of the organic component? What is the role of the background aerosol? How big fraction of the mismatch can be attributed to errors in calculating the time shift? The authors

C15211

seem to suggest numerical diffusion to be a reason for this mismatch; if so, what would be the implications for measurement data?

Since the original submission of the article the research of this topic has advanced to the point that we decided to remove the time shift analysis from this article.

Specific comments:

Page 18792: "The effects of numerical diffusion were not considered in this study." Can you comment on the expected importance of numerical diffusion and what factors that would influence the error induced by it?

The level of numerical diffusion in the simulations is actually negligible (compared with effects of omitted processes, experimental uncertainties in real measurements etc.). Thus we removed this unnecessary and potentially confusing statement.

Page 18788, equation 5 and related discussion: Can the author explain why GR around 3-7 nm is used?

This is in accordance with the previous studies, who have decided to use this diameter range, probably due to it being relatively simple to estimate from measurement data. The MS now states: "... it is easier to estimate an average growth rate from the rate of change in mode diameter of particles from a wider size range, e.g. (roughly) 3–7 nm [Sihto et al., 2006; Riipinen et al., 2007]."

Page 18791: Equation 8 is a suggested improvement over the analysis of J3. It is however not clear to me if this improvement is the result of trial and error, or if it is based on physical considerations. The text is a little bit unclear in this perspective. Just tuning in the equation using different size ranges does not have any real implications for treatment of measurement data. How will these assumptions hold if the resolution of the instrument is changed, as example?

Intuitively, the number concentration of 6 nm particles is better estimated by using a range of 5-7 nm instead of 3-6 nm. One could certainly fine tune the size range to get

C15212

more exact correspondence, but this one is simple enough to be applied in practice. The sentence introducing Eq. 8 is now mildly rephrased to be more intuitive: "... a better solution would be estimating n_6 from a size range closer to 6 nm, for example...".

Page 18791, line 20: "...most of the deviation between Eqs. (3) and (5) occurs because of coagulation...". What do the authors mean by this, is it that the calculation of coagulation is wrong? Try changing to "...is caused by..." or "...occurs due to error in the estimate of the coagulation term..." Furthermore, doesn't the result in table 1 (BKG→0) indicate that coagulation is completely responsible for the deviations? Or is it also connected to the CS? Please comment on this.

The sentence is now rephrased as suggested by the reviewer. While the results in table 1 suggest "perfect" agreement if no particle background is present, Eqs. 3 and 5 are still not the same, albeit very close (cf. difference in diameter range considered).

Page 18792, line 21-22: Please clarify what you mean here.

The chapter on time shift analysis was removed from the revised manuscript.

Page 18792, line 6-7: Do not get what the authors mean by phenomena here. Do we mean processes, size ranges or what?

The words "aerosol dynamical" are now added: "Nevertheless the trends related to and approximate magnitudes of different aerosol dynamical phenomena should be general."

Minor comments:

Page 18788, line 5: "...basing on them..." → "...based on..."

The reviewer is probably referring to line 15 on page 18787. Rephrased to: "...from the number concentration data."

Page 18789, line 17-18: "For an example. . ." → "As an example. . ."; "For example. . ."

C15213

Rephrased to: "For example..."

Page 18789, line 18: Can a background be "more abundant"? Sounds awkward. In fact, the whole sentence needs rephrasing as it is currently hard to understand what the authors really mean.

The sentence is split to shorter, clearer sentences: "For example, a higher concentration of pre-existing (background) particles would amplify the coagulation sink. Consequently, the number concentration of 3–6 nm particles would decrease, affecting all terms in Equation 5. However, due to the more direct effect of the pre-existing particles on the coagulation sink, the coagulation term would increase the most."

Page 18790, line 7: "We modified the analysis to be more accurate. . ." sounds a little bit subjective. Maybe should write "We performed more detailed calculations. . ."

Agreed. Rephrased as suggested.

Page 18790: Line 10: ". . .comparing these. . .", should be stated more clearly what you are comparing here in order to improve readability.

Rephrased to: "By comparing these coagulation sinks..."

Page 18791, line 1-5: Please clarify.

The part is now rephrased: "... our sensitivity test suggests that the applied particle diameter range affects the accuracy of J3 significantly, with a broader range giving greater accuracy, until 3–13 nm, after which the applied Eq. 5 begins to underestimate the formation rate. Of course, this is caused by a numerical effect: the significant increase in the denominator of the growth rate term in Eq. 5 decreases its value. While increasing the level of agreement between the two equations for formation rates, the difference caused by the coagulation term remains. Obviously the exact numbers presented in Table 2 will vary depending on the case..."

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 18781, 2010.

C15214