

Review of the manuscript:

In their manuscript, “Using a combined power law and log-normal distribution model to simulate particle formation and growth in a mobile aerosol chamber”, M. Olin, T. Anttila and M. Dal Maso have described a novel method for simulating the dynamics of aerosol particles, with the emphasis on simulating the early growth of a freshly-formed particle mode in a computationally cost-efficient manner. The authors have tested their model against previous aerosol dynamics models in a few simplified test scenarios to estimate the accuracy of the novel model, and also to demonstrate both the computational efficiency of the novel model in comparison to more accurate models and the accuracy of the novel model in comparison to other models with similar computational burden. Finally, the authors have used the model to reproduce a new particle formation and growth event as observed in aerosol chamber measurements. The main conclusions of the paper are that the newly-developed model is able to provide concentration, surface area and mass concentrations within a few percent to those obtained with a highly accurate model, and that the novel model is able to represent simultaneous new particle formation and particle growth, which is beyond purely log-normal model, which is often used for cost-efficient representation of an aerosol population.

Based on the results presented in the manuscript, the new model seems like a useful compromise between accurate and computationally cost-efficient representations of a particle population, and the manuscript is in the scope of ACP journal. A more thorough evaluation of the model is needed, however, and the language of the manuscript should be revised.

General comments:

The main objectives of the manuscript are: 1) to describe the new model in detail, 2) to evaluate the accuracy and computational cost-efficiency of the model, and 3) to demonstrate the applicability of the model using a real life example. I find that the first objective is covered quite well in the manuscript. The second objective, however, is not covered sufficiently: the new model is evaluated against more accurate models in a handful of scenarios, but a more thorough examination is needed. The main issue is that only size-independent growth rates are considered. Another issue is the parameter γ , which is left as a free parameter in the model, but relatively little consideration is given to how sensitive the model results are to the choice of γ , or, especially, how to choose the value of γ for given simulation conditions. These, and some other issues, are described in more detail in the “Specific comments”, below. Regarding the third objective, a single example of reproducing the time evolution of a particle size distribution during a new particle formation and growth event as observed in a chamber experiment is provided. A few different examples would probe the capabilities of the model much more comprehensively. Even more problematic, however, is that in this example, the particle formation and growth rates representing the measurement conditions are obtained with inverse modelling using another, more accurate, model, and those values are used as input in the novel model with the results being compared to the results from the more accurate model. In other words, the only connection to the real life measurement is that the formation and growth rates used as inputs in the models represent the measurement conditions, but otherwise there is not much difference to the scenarios used to evaluate the model. A more suitable demonstration of the usage of the model would be, for example, to estimate the formation and growth rates

representing the measurements using inverse modelling with both the accurate model and the novel model, and then compare those values. Finally, the language of the manuscript is not very good at times. The grammatical errors are not exhaustively listed in the “Technical corrections”, and a greater care for punctuation, for example, would be needed.

Specific comments:

1. The title of the manuscript implies that the new model is able to simulate the actual process of particle formation, which is misleading, as the formation rate is used as an input in the model, and also that the emphasis of the manuscript would be on simulations related to the mobile aerosol chamber, but only one example is given. The title should be revised.
2. On page 7, lines 19-23, it is stated that the mass growth rate is assumed to be proportional to D_p^2 in the model, even though only one of the three conditions justifying that assumption is met. Then, on page 8, line 1, it is stated that this assumption of proportionality results to condensational growth rate being size-independent. The growth rates observed in the atmosphere, however, are not size-independent for the particle size range considered in the model (e.g., Kuang et al., 2012). Furthermore, in all of the simulations used to evaluate the new model, the growth rate is assumed to be size-independent. In other words, the model includes an approximation that is in contradiction to observation, but the error caused by this approximation is not probed at all when evaluating the model accuracy. This is a considerable omission, and additional simulations with size-dependent growth rates should be added. The sectional models used as reference obviously do not suffer from this approximation.
3. As stated on page 16, lines 7-14, the size distribution observed in the chamber measurement is combined from data from three separate instruments, but there is no information to how they are combined. For example, the information from all the three instruments could be used simultaneously (e.g., Viskari et al., 2012) to provide the size distribution, but this seems unlikely based on Figure 4. Another option is that the raw data are inverted separately, which raises the question, how a $dN/d\log D_p$ values are calculated for PSM and CPC, which only measure total concentration above a given cut diameter? Without the information of how the observed distribution is obtained, one cannot properly evaluate the accuracy of the new model against those measurements, especially in the sizes below the range of SMPS measurements. A proper explanation of how the measured size distributions are obtained should be given.
4. Beginning from page 13, line 6, until page 14, line 3, it is described how the particles are transferred from PL distribution to LN distribution due to condensational growth. Based on Figure 10, it seems that the value of D_2 is affected by the choice of γ , so this relation should be given explicitly. Furthermore, it is inconvenient for any practical use of the model that γ is a free parameter, but very little information is given as to how to choose that value for given conditions to be simulated. In the manuscript, only one example case is provided, which makes it difficult to assess if the optimal value of γ for that case can be generalized to other cases or not. For example, does the optimal value of γ depend on the particle growth rate or formation rate? In order to facilitate any future use of the model, the authors should provide some advice for choosing the

value of γ , preferably with added examples of comparisons to measurements with different conditions.

5. Page 1, lines 12-16: the first paragraph of the manuscript feels like a few bullet points collected to give some background, and then the rest of the introduction deals with various approaches to modeling particle size distribution. It would serve the reader to have a little longer and more coherent description of the framework and motivation of modelling size distributions.

6. Page 5, line 27: Is the intramodal coagulation really the only process initiating the formation of the LN distribution, or can the condensational transfer initiate it also? According to Eq. (50) the amount of condensational transfer does not depend on the LN distribution, or on the existence of one. Furthermore, on page 17, line 10, it is stated that the coagulation transfer was neglected when simulating conditions of chamber experiment, but LN distribution is still seen in the results. This needs to be clarified.

7. Page 7, lines 5-11: Does this part have something to do with the current work? As far as I understand, the growth rate is used as an input value in the model in all of the simulations, and the condensation process is not really simulated. If this paragraph is important, then it should be made clear why, and if not, it would clarify the article to remove it.

8. Page 7, lines 12-14: It is unclear why the parameters in Eq. (16) need to be considered to vary with t and D_p for the mass growth rate to vary with t and D_p . According to Eq. (16), the mass growth rate depends explicitly on D_p , and if any term depends on t , then the mass growth rate should depend on both t and D_p .

9. Page 7, lines 23-25: I do not understand why the new particle formation rate is in this sentence, please clarify.

10. Page 10, lines 18-20: It is stated that the degrees of quadratures are low, but it would serve the reader to provide some examples, how much the simulation results would change due to lower or higher degree of the quadratures used in the model.

11. Page 14, line 16: It is stated that the diameter of newly-formed particles was assumed to be 1.6 nm. Would the results and/or conclusions change with another choice of this diameter? If so, it should be presented, and if not, then that should be mentioned.

12. Page 15, lines 22-23: What is the reason for only considering coagulation transfer, but not condensational transfer?

13. Page 16, lines 13-14: It would serve the reader to explain with a few words how the EEPS and ELPI+ are used to ensure the stability of the aerosol distribution.

14. Page 16, lines 28-30: The PL+LN model was used to estimate $J(t)$ and $g(t)$ via inverse modelling, before those estimates were fine-tuned with FS model. Comparison of the best estimates of J and g from inverse modelling using both the PL+NL and FS models would be an excellent way to demonstrate the capability of the new model. Adding such comparison would increase the

practical use of the manuscript. Performing such comparison manually might not be the most robust approach, though.

15. Page 17, line 15: Only the distributions at the end of the simulations are shown, but it would serve the reader to also provide an example in which the distributions from the PL+NL model and FS1000 model would be compared at other times. This could be done, for example, by showing a surface plot of the relative difference between the two as a function of time and diameter. Such plot would make it possible to evaluate the accuracy of the PL+NL model at all particle sizes and stages of a new particle formation event, instead of just the end distribution.

16. Page 18, lines 31-32: Even if the resolution of the measured distribution is poor below the size-range of the Nano-SMPS measurement, the measured GMD and GSD values would provide a valuable comparison to those from the simulations, especially towards the end of the time domain. I suggest showing also the measured GMD and GSD values in Figures 8 and 9, respectively.

17. Page 19, lines 1-6: This paragraph is a little confusing. I understand that the inverse modelling using the most accurate model produces the best estimates for the $J(t)$ and $g(t)$ representing the actual measurement conditions, and those values are then used as input in the other models, instead of some other values of $J(t)$ and $g(t)$ that would produce better correspondence between those simulation results and measurements. Choosing against what these simulation results are compared to, however, depends on the motivation of the comparison: If the point of interest is, how similar are the distributions simulated with a simple model and a more accurate model, when the same $J(t)$ and $g(t)$ values are used as input in both models, then the distributions from those models should be compared against each other. On the other hand, if the point of interest is, how well does the simple model reproduce the measured size distribution, when the best estimates for $J(t)$ and $g(t)$ representing the measurement conditions are used as input in the model, then the comparison should be against the measurement data, not the accurate model. The authors should revise the paragraph and make it clear what they want to say with it.

Technical corrections:

1. Page 1, line 4: the word “validate” refers to something being labeled as valid, which is not really a proper metric in case of an aerosol dynamics model. I would suggest changing it to “evaluate” here and on other instances the word “validate” is used.
2. Page 1, line 22: D_p and t are used, but they have not been defined yet.
3. Page 2, line 1: there should be a comma on both sides of “e.g.”. This issue occurs repeatedly in the manuscript.
4. Page 2, line 4: should read “in which” instead of “of which”.
5. Page 2, line 5: consider changing “changed” to “are allowed to vary”.
6. Page 2, line 5: One should avoid starting a sentence with “however” when the meaning is “nevertheless”. The same issue occurs repeatedly in the manuscript.

7. Page 2, line 10: it is unclear what “too” means in this context.
8. Page 2, line 14: consider changing to “...the effect of numerical diffusion to their results is unknown.”.
9. Page 4, line 2: consider using a comma on both sides of the defined variables.
10. Page 4, line 13: there should be a comma on both sides of “i.e.”. This issue occurs repeatedly in the manuscript.
11. Page 5, line 15: consider changing “represents” to “presents”.
12. Page 6, line 11: consider changing to “where terms on the right hand side denote...”.
13. Page 7, line 2: it would serve the reader to clarify that “*i*” is just a shorthand notation for either PL or LN.
14. Page 7, line 5: variables longer than a single letter should not be in italics, “Kn”. This error occurs multiple times in the manuscript.
15. Page 7, line 6: add “where” to the beginning of the line.
16. Page 7, line 21: the word “latter” is confusing in the case of more than two items.
17. Page 14, line 15: the FS models are referred to in plural while only a single model was mentioned earlier in the paragraph, please clarify.
18. Page 16, line 1: the “254 nm” is probably the UV wavelength, but it should be made clear.
19. Page 16, lines 30-32: please revise the sentence, it is grammatically incorrect.
20. Page 16, line 33, until page 17, line 1: This sentence is grammatically incorrect, and also somewhat ambiguous about how the deposition coefficient was obtained, which needs to be made clear.
21. Page 17, line 16: should read “equal to”.
22. Page 17, line 18: should read “the highest”.
23. Page 17, lines 24-25: I eventually understood it, but this sentence is quite confusing, consider revising.
24. Page 18, line 15: consider changing “represents” to “presents”.
25. Page 18, lines 28-30: I understand the message, but the sentence is poorly worded, consider revising the sentence.
26. Page 19, line 7: consider changing “represents” to “presents”.
27. Page 19, lines 8-9: consider changing to “Conversely, using a high value of γ produces a more log-normal like form...”

28. Page 19, lines 21-22: "...obtaining input parameters as the model output through inverse modelling" is quite unintuitive way of saying "... using inverse modelling to obtain the best estimates for parameters used as input in the model". Consider revising.
29. Page 19, line 31: The word "conservation" is misleading in this context, as it is commonly used to describe what parameters are conserved in a model, instead of how similar certain parameter values in results from different models are.
30. Page 20, line 8: Saying "simple log-normal distribution model output GMD relatively well" seems to imply that there were rarely any issues with getting the GMD value from the model. Consider revising.
31. Page 23, Table 1: The value of the deposition coefficient is given in nm/s in the text, but in nm/h or nm/s in the table. Using nm s^{-1} consistently is advised.
32. Page 24, Table 3: The first sentence "Computational costs and relative errors of different models using the chamber data." is problematic as the table shows errors in certain model outputs, not in the models themselves, and, furthermore, no chamber data per se was used in the simulations, only some input values representing the conditions of the chamber experiment.
33. Page 25, Figure 1: The y-label refers to size distribution as "dN/dlogDp", while "dN/dlnDp" is used in the equations. Consistent notation is suggested to avoid confusion.
34. Page 30, Figure 6: It is often a good practice to provide ticks for the minimum and maximum values of both the x- and y-axis in figures, as it facilitates more accurate comparison to the presented values. In this case, for example, the reader might want to reproduce the model described in this manuscript, and then compare the results from that model to results presented in the manuscript using the same input values.
35. Page 31, Figure 7, caption: It should read "...after the UV lights were switched on...".

References

- Kuang, C., Chen, M., Zhao, J., Smith, J., McMurry, P. H., and Wang, J.: Size and time-resolved growth rate measurements of 1 to 5 nm freshly formed atmospheric nuclei, *Atmos. Chem. Phys.*, 12, 3573-3589, doi:10.5194/acp-12-3573-2012, 2012.
- Viskari, T., Asmi, E., Virkkula, A., Kolmonen, P., Petäjä, T., and Järvinen, H.: Estimation of aerosol particle number distribution with Kalman Filtering – Part 2: Simultaneous use of DMPS, APS and nephelometer measurements, *Atmos. Chem. Phys.*, 12, 11781-11793, doi:10.5194/acp-12-11781-2012, 2012.