Retrieval of process rate parameters in the general dynamic equation for aerosols using Bayesian state estimation of aerosol dynamics: BAYROSOL1.0

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1 Thanks

We thank the reviewer for his/her thoughtful comments and will revise our manuscript accordingly. In the following are our point-by-point responses to the reviewer's remarks in such a way that we have listed the reviewer's remarks in blue and our reply is in black font.

2 General comments

2.1 Paragraph 1

It seems odd that there is no source term for nucleation in equation (1). I see that it is included as a boundary condition on particle flux in, but even with that it seems like these equations do not correctly represent particle nucleation and growth. Since if the net growth by condensation of d_p^{min} is set to J, then either there is no growth of d_p^{min} to larger sizes or the loss of d_p^{min} is included in J.

The reviewer is correct in noting that in equation 1 there is no term for nucleation and we have explained our approach insufficiently. Our approach here considers particle dynamics above the size range in which the actual nucleation occurs, i.e. a typical DMPS or SMPS measurement range. Then, the appearance of new particles to the lowest end of the measurement range occurs through condensation from even smaller particles that have nucleated slightly earlier. This process has sometimes been called "apparent particle formation", and we adopt this terminology here to avoid further confusion. Such a growth of particles across the lower limit of our particle size range is most conveniently treated mathematically as a boundary condition, resulting in the discretized model (equation 5) as a source term for the lowest size bin. Thus, in our revision we will replace "nucleation" with "apparent particle formation" everywhere and add on page 4 (before equation 2): "*We do not include an explicit nucleation term in equation 1 as we are considering a size range typical for particle mobility (DMPS*

or SMPS) measurements which is above nucleation size. Then, appearance of new particles to the measurement range occurs by condensational growth of freshly nucleated particles from below measurement range. This process is sometimes called apparent particle formation (e.g. Lehtinen et al., 2007) and mathematically it is conveniently treated as a particle concentration flux in size space ($cm^{-3}s^{-1}$) boundary condition for the GDE." This flux term can be written as a product $g \times n$ (equation 2), signifying that it is affected only by the condensation. This, however, does not mean that the deposition is neglected in the smallest size class.

Additionally, J is defined as flux of particles (number of particles per area per time), but then is referenced as particle concentration rate (# $cm^{-3}s^{-1}$) later in the discretized model and numerical simulation. In the discretized model there is in fact a nucleation and growth term for the first bin, so it seems like the error is in the representation of the continuous GDE in equations (1) and (2). This needs to be corrected or clarified. Also, in equation (1) d_0 is used as the lower limit integrated over for coagulation sink but d_0 is not defined.

This comment by the reviewer likely stems from our poor explanation of the treatment of nucleation (or apparent particle formation, see our reply to previous comment). The discretization of the GDE with apparent particle formation rate as a boundary condition results directly in the discretized equations 5 and 6, so no consistency problem there. We agree with the reviewer that we use the term flux non-traditionally as the propagation of particle concentration in our case occurs in particle diameter space (and not "normal" space). This is clarified by the addition mentioned in our response to the previous comment:"*We do not include an explicit nucleation term in equation 1 as we are considering a size range typical for particle mobility (DMPS or SMPS) measurements which is above nucleation size. Then, appearance of new particles to the measurement range occurs by condensational growth of freshly nucleated particles from below measurement range. This process is sometimes called apparent particle formation (e.g. Lehtinen et al., 2007) and mathematically it is conveniently treated as a particle concentration flux in size space ($cm^{-3}s^{-1}$) boundary condition for the GDE*" In the revised version, we will also define d_0 clearly. It is the actual (physical) diameter at which the nucleation occurs. Moreover, we have highlighted the difference between d_0 and $d_p^{\min}$ – the latter of which is the "apparent nucleation size", which represents the smallest diameter in the model. Also, we will change the coagulation source in equation (1) since the integral range should start from the same size as the coagulation loss, d_0 instead of 0.

2.2 Paragraph 2

It is noted that Case 1 & 2 are set-up to study estimation stability to see if the method can estimate time-invariant wall loss even though the loss rate follows a 1st order Markov model. However, the estimated loss rate is only shown at one time and it is not discussed further. Did the estimated loss rate vary over time, and by how much?

Yes, the wall loss was treated as time varying, even if it is expected to be time invariant (but size dependent). The Extended Kalman filter (EKF) results for the wall loss rates showed time dependency while the Kalman smoother (FIKS) produced very weak time dependence. See some results at different times in fig. 1. We choose not to show these figures in the

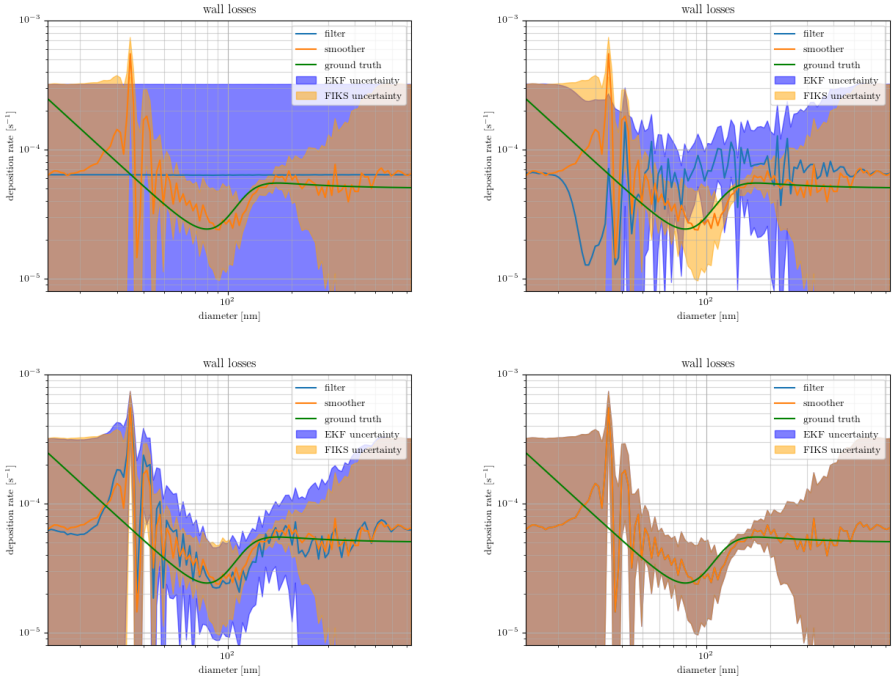

Figure 1: Wall loss estimation for four different instants in time.

manuscript but will add a sentence on this in the revised discussion.

2.3 Paragraph 3

What is the range in SNR between cases 1 and 2 as well as between cases 3 and 4? How is SNR adjused?

The SNR was adjusted by choosing the sample volume (V, via the detector-sample-flow rate ϕ_a) of the CPC. As noted in Section 3.1.1, the SNR of CPC data increases with V. The associated Poisson distributed noise is the main source of noise also in the real experiment, and choosing

V is a trade-off between SNR and the duration of the measurement. In the studied cases, the SNR is controlled by ϕ_a so that the time base of the measurements is the same between different SNRs. The actual ranges for the SNRs are case 1 $[0, 6426]$, case 2 $[0, 64.26]$, case 3 $[0, 4440]$ and case 4 $[0, 44.4]$, and we will mention them in the respective sections.

2.4 Paragraph 4

The observed difference in estimated and true nucleation rates in cases 3 & 4 is quite interesting. It seems like perhaps the nucleated mass rate matches closer than the nucleated number. Is this the case? If so, it would be interesting to note that FIKS can recover the nucleated mass rate when there are uncertainties in the nucleated particle size.

The underestimation of nucleation rates in cases 3 and 4 resulted from the fact that we were comparing particle formation rates at two different sizes, 0.87 nm and 1.1 nm. The rates at 1.1 nm are lower because as the particles grow from 0.87 nm to 1.1 nm their concentration is decreased by (mainly) deposition onto the walls. We have now corrected our analysis and in figure 2 one finds the new versions of figures 6b and 8b, showing an excellent match for the predicted particle formation rates. We also change the text (on page 19) accordingly.

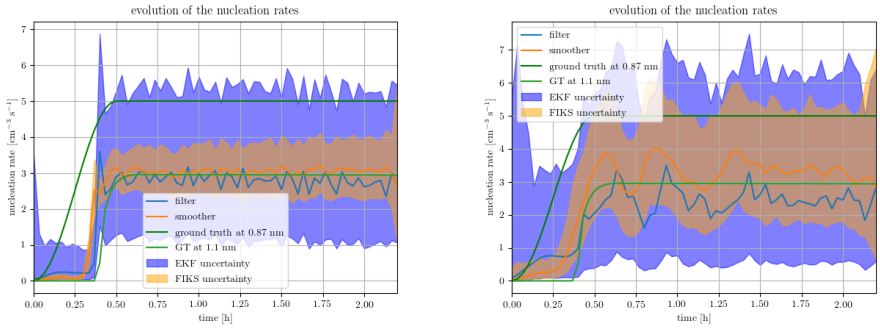

Figure 2: Adding the true (simulated) particle flux at 1.1 nm (size at which the nucleation is estimated by the method), a.k.a. apparent nucleation rate.

3 Minor corrections and suggestions

1 Line 31-32 "...paying also attention to the uncertainties" is confusing wording. Maybe change to "analyze the data with care and pay attention ..." We will revise this as suggested.

2 In general, the citations should have the format (Author1, year1; Author2, year2; ...) unless the citation is a subject in your sentence in

which case the format should be just the year in parenthesis, i.e., "this thing was described by Author1 (year1) and Author2 (year2)" We will revise the citations according to the guidelines of the journal.

3 GR needs to be defined as growth rate when it is introduced in line 38. This will be corrected in the revised version of the manuscript.

4 Citations are repeated in the paragraph starting at line 60. This needs to be fixed. This will be corrected in the revised version of the manuscript.

5 Line 70 change to "the Bayesian approach was adopted to estimate aerosol size distributions" This will be corrected in the revised version of the manuscript.

6 Line 182 - describe what the notation $]0, 1[$ means. I am used to seeing $x \in (0, 1)$ for $0 < x < 1$ and $x \in [0, 1]$ for $0 \leqslant x \leqslant 1$

7 It would be helpful to explicitly describe what $\tilde{y}_i^k$ and z_i^k represent (number of particles counted?). z_i^k is defined in equation (16) and the line above it and $\tilde{y}^k$ is the number of particles counted by the CPC, which we add after equation (17)

8 Figures 7a - 7d need timestamps. Will be done. And the same will be applied to figures 9a – 9d.

9 Reference Appendix B in the text (near lines 180-190) to describe how r_ϕ is chosen. This will be corrected in the revised version of the manuscript.

10 In algorithm 2, it seems like it should be a loop over $k = K - 1, \ldots, 1$ or there should be a separate case for if $k = K$ since it is not clear that $\Gamma^{K+1|K}$ or $X^{K+1|K}$ exist. Thank you for noticing this mistake. Will be corrected.

11 Figures 2a and 2b look very similar to my eyes. It would be nice to show the surface plot of their difference, potentially instead of the current figure 2b. The difference in the transfer functions is resolution, which is illustrated in the panel c) of figure 3 for one channel. The transfer function used in the method corresponds to the average of the "true" transfer function over each discretization bins, hence the difference in amplitude between the fine and coarse models. It is clearly illustrated in the panel c) of figure 3 where the maximum value of the averaged model, in orange, is smaller than that of the fine model, in green — which is merely the evaluation of transfer function, and not the average. The figure 3 will replace figure 2 in the manuscript.

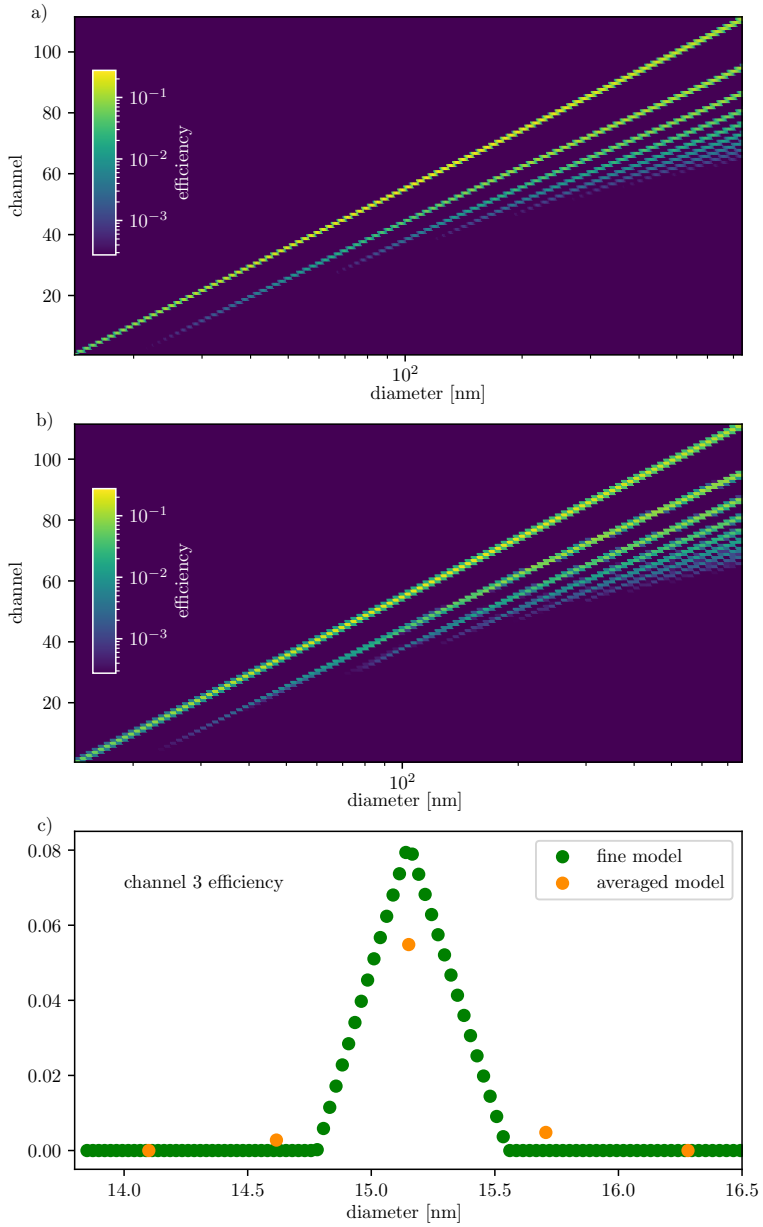

Figure 3: New transfer/kernel function plot.

Additional reference

Lehtinen, K. E. J., Dal Maso, M., Kulmala, M. and Kerminen, V.-M. (2007) Estimating nucleation rates from apparent particle formation rates and vice versa: Revised formulation of the Kerminen-Kulmala equation. Journal of Aerosol Science, Vol. 38, No. 9, 2007, p. 988-994.