# 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 Comments

### 2.1 Comment 1

If I understand correctly, no constraints for the fitted parameters are used in the preset work. Could a measurement of the gas-phase condensable vapors provide a useful upper-limit constraint for the condensation rate parameter g (corresponding to irreversible condensation)? Would this improve the results? Similarly, the upper limit of the nucleation rate could possibly be assessed if the identities of the nucleating vapors are reasonably well known (at least in well-controlled laboratory experiments).

The positivity constraint is the only constraint applied. If additional physical information exists, e.g. vapor concentrations as the reviewer suggests, this can be straightforwardly incorporated in the estimations. And, as a general rule, the more information available, the better the estimation.

### 2.2 Comment 2

Figure 1: A minor observation on the NE event: the pre-existing distribution of larger aerosols seems to persist throughout the event, although normally atmospheric observations show that it is diluted due to boundary layer growth around noon (thus also enhancing the particle formation efficiency). Can such a dilution effect be added to the Bayesian model equations?

Yes, dilution can be straightforwardly added to the model, as can be any process that can be modeled with a time evolution equation.

### 2.3 Comment 3

P7, L173: It is reasoned that all the unknown parameters of the GDE are non-negative. However, isn't it possible that particles shrink at low vapor concentrations (e.g. Salma et al., Atmos. Chem. Phys., 16, 7837-7851, 2016), thus making the condensational growth rate g negative? (Similarly, also the formation rate J may be negative in the case of shrinking particles.)

Yes, an aerosol can also evaporate, causing a negative growth rate. And as the reviewer points out, this makes also a negative apparent particle formation rate possible. The choices made in this paper (the parametrization, i.e., the softplus variable transform in Equation (10) and the way we discretize the GDE in Equations (5)-(6)) imply that the growth and formation rates are positive. However, the adaptation to cases where aerosols evaporate is straightforward.

We add the following sentences in the revised manuscript, after equation (6): "The choice for approximating the derivative in the growth term in the discretization of the GDE is made here assuming that the particle growth rate is positive. The modification to cases where the aerosols evaporate, i.e., where growth rate is negative, is straightforward."

### 2.4 Comment 4

Is it controlled that the applied Euler's method for time integration does not contain (cumulative) errors? While the integration is stabilized by the CFL criterion, does it ensure that a shorter time step will not have a notable effect on the result? Short steps may become relevant at high particle concentrations in polluted atmospheric environments, such as very polluted urban conditions.

We acknowledge that the explicit Euler's method used for time integration is not the most accurate one. However, the adoption of any other time integration scheme (such as implicit Euler or Crank-Nicholson) is straightforward, and only affects the evolution model. On the other hand, in cases where the state-space systems are identifiable, the state estimates are generally tolerant to inaccuracies in the evolution model: the sequential measurement data is used for correcting the inaccurate predictions by the evolution model at every time step, and this prevents the cumulation of the time discretization error. We also note that the tolerance with respect to such modeling errors can be further improved by so-called approximation error analysis J. Huttunen and J. Kaipio (1). We will add a sentence on this topic also to the manuscript.

### 2.5 Comment 5

While previously introduced GDE-based methods to assess particle growth rates or other parameters (i.e. those cited on P3, L60-67, and also the more

recent methods proposed by Pichelstorfer et al., Atmos. Chem. Phys., 18, 1307-1323, 2018) do not include parameter uncertainty estimates, can it still be useful to test also these methods against the Bayesian state estimation?

We have chosen here not to compare against other methods as the use of simulated measurement data enables us to compare with the true 'answers'. BAYROSOL is, however, compared against just the methodology mentioned by the reviewer Pichelstorfer et al. (2) (Pichelsdorfer et al., 2018) in a forthcoming paper (submitted to Atmospheric Chemistry and Physics) where the method is applied to experimental aerosol size distribution evolution data obtained at CLOUD/CERN. The methods agree very well, however, as mentioned, BAYROSOL also provides uncertainty estimates for the parameters estimated.

### 2.6 Comment 6

It is explained that the evolution of the aerosol size distribution is determined by the nucleation, condensation, coagulation, and sink parameters. I'm wondering if the aerosol particles can exist in different charging states, and if this can have effects on the distribution through e.g. modified growth and coagulation kernels?

Yes, different charging states are possible, as suggested by the reviewer, and charges will definitely affect the dynamics. Here, in our first tests of BAYROSOL, we have, however, started from the simplest case — neutral particles. It is, however, conceivable that the methodology applies well also to an aerosol population with several charging states if measurement of both size and charge distributions of the charged particles are available. Then, of course, GDEs for the charged particles are needed, as e.g. described in Leppä et al. (3).

### 2.7 Comment 7

P4, L93-94: It is stated that the formation of particles occurs typically at 1.5-2 nm. Yet, the nucleation size in the simulations is assumed to be only 0.87 nm, which is barely a molecular size and would generally be expected to correspond to "pre-nucleation" molecular clusters. How was this size chosen? Are the results affected if the size is erroneously assigned?

The size 0.87 nm was chosen arbitrarily, and as the reviewer remarks, is unphysically small if atmospheric nucleation is concerned. As the purpose of this manuscript, however, concerning nucleation is to test how well the nucleation rate is estimated, the actual choice of nucleation size does not affect any of the results concerning method performance.

### 2.8 Comment 8

Furthermore, for the steady state (SS) simulation case, the modeled size range is divided to 1731 logarithmically distributed size bins in the range [0.87 nm, 10.00 nm] (P16, L383). For such a high number of bins in a narrow range, the bins are very dense especially at the smallest particle sizes.

Is it ensured that the width is still physically consistent, that is, corresponds to at least one-molecule increment in terms of particle growth? I believe that for large atmospheric molecules, such as oxidized organic species, 10 nm-particles may consist of significantly less than 1731 molecules, in which case the bin widths would be unphysical.

The reviewer is correct in stating that the bin width, especially at the lower end of the size spectrum, is unphysically narrow if compared with molecular size. Mathematically, this is, however, no problem as we are numerically solving the continuous form of the GDE.

### 2.9 Comment 9

P4, L96-97: It is stated that the particle flux to the smallest measurable size class is driven by condensational growth, as defined by Eq. (2). Can coagulation also play a role in polluted environments with high concentrations of nucleated particles? Would this affect the model formulation?

Yes, in the most polluted environments the particle number concentrations can be so high that coagulation can play an important role in the particle flux into the measurable size range. In such case, the boundary condition described by Equation (2) may no longer be valid. However, in this case, we can include the flux of particles caused by coagulation into term J, and the form of the discretized GDE will remain unchanged.

### 2.10 Comment 10

P16, L361 onward: Why is a wall loss rate estimate discussed for the "nucleation event" (NE) case? It is explained that the NE case represents a typical particle formation event in the atmosphere. Which type of walls in the ambient atmosphere does the loss rate correspond to, or does it refer to the walls inside a measurement device?

What is meant by wall loss is actually a deposition loss, e.g. sedimentation. We will make use of the term losses instead of wall losses to avoid any further confusion. The measurement model does not account for losses in the tubing of the device, though it can be straightforwardly modified to take it into account if the loss rates are known.

# 3 Technical comments

## 3.1 Comment 1

The colored solid lines, corresponding to the "true" and estimated parameters as functions of time or particle size, in most figures are somewhat difficult to see; can they be made e.g. thicker and brighter (and maybe have different line styles)?

In the revised version of the manuscript, we will increase the linewidth parameter of the plots to thicken the curves.

Also, the shaded areas corresponding to the uncertainties in the estimates could preferably be lighter and/or more transparent in order to see the lines better. It may also not be obvious by first look that the brown shade is the overlap between the blue and yellow shades; the figure legends list blue and yellow shades, but in most figures the yellow is almost entirely missing and there is only brown / blue.

The figure will be modified to make the blue and orange shaded area more transparent.

## 3.2 Comment 2

P2, L42: Please remove the comma in "Methods 1 and 2 are applicable to cases, in which. . ."

We will modify the punctuation to restore the proper grammatical sense of this sentence.

# References


[1] J. Huttunen and J. Kaipio *Approximation error analysis in nonlinear state estimation with an application to state-space identification*, Inverse Problems, 23, 5, p 2141, 2007.

[2] Pichelstorfer et al. *Resolving nanoparticle growth mechanisms from size- and time-dependent growth rate analysis*, Atmospheric Chemistry and Physics, 18, 2, 1307-1323, 2018.

[3] J. Leppä et al. *Ion-UHMA: a model for simulating the dynamics of neutral and charged aerosol particles*, Boreal Environment Research, 14, 559, 2009