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Interactive comment on "Novel estimation of aerosol processes with particle size distribution measurements: a case study with TOMAS algorithm" *by* Dana L. McGuffin et al.

Anonymous Referee #1

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Review of gmd-2020-281: "Novel estimation of aerosol processes with particle size distribution measurements: a case study with TOMAS algorithm"

General comments

In this work the authors present and examine a proposed new method for incorporation of information from aerosol particle size distribution measurements into atmospheric chemical transport models (CTMs). Specifically, the method utilizes an inverse modeling algorithm to constrain and adjust estimates of relevant aerosol microphysical process rates based on changes to the size distribution and the use of the TOMAS aerosol microphysics model. Aerosol nucleation, emissions, and growth processes are

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Discussion paper

investigated here using a simple “box model” to assess the utility and efficiency of the method. The stated goal of the work is to investigate if such a method could potentially be utilized in a 3D CTM to better constrain estimates of such aerosol process rates.

The manuscript is generally well written and understandable, and includes reasonable first steps to investigate the utility of the method. The authors select three “inventory variables”, which can be derived directly from measured size distributions, and use the inverse model to link changes to these variables with changes to the various aerosol process rates. In using synthetic measurements (including reasonable estimates for measurement noise), they demonstrate that under conditions in which only the investigated aerosol processes are relevant to changing size distributions, the method can successfully estimate the various underlying process rates. When applying the method to actual observed measurements the estimates of process rates expected to be sensitive to additional atmospheric and aerosol processes are less successful, though this is expected for the simple box model scenario. The result is a successful demonstration of the potential of the method, though it remains to be seen (in future work) if information from actual size distribution measurements can be incorporated into a 3D CTM where more degrees of freedom may make inverse modeling more difficult.

Extraction of information from aerosol particle size distributions can be particularly difficult as many underlying processes—not all of which are well measured in the actual atmosphere or well represented in idealized CTMs—contribute to changes in both the aerosol particle size distribution and the derived inventory variables used here. As such, significant caution is warranted when assessing efforts to do so. However, efforts to include better estimates of more fundamental aerosol process (such as the nucleation, emission, and growth rates investigated in this work), as opposed to variables that serve more as proxies for actual aerosol processes (such as aerosol species mixing ratios, as noted by the authors), are valuable contributions to aerosol modeling efforts. In this regard, the authors may in fact be underselling the potential of their results to some extent, as computationally efficient methods to better incorporate such

fundamental aerosol processes into 3D CTMs is both important and valuable.

As such, I recommend the manuscript be published in Geoscientific Model Development subject to minor corrections and consideration of several questions I list below.

Specific comments

Several related open questions remained that appear to not be fully addressed within this work. While they are likely beyond the scope of this manuscript and need not be fully resolved here, it may be worthwhile for the authors to include a discussion of them for efforts moving forward with this method.

1. The question of the relevance of other aerosol processes on process rate estimates from the inverse model.

In particular, a set of measured aerosol particle size distributions in the actual atmosphere will be subject to variation from a wide variety of processes. Some of these can be simulated by either the CTM or TOMAS to varying degrees of accuracy while others may not be included at all. As a first step the authors select three aerosol processes for their relevance to prediction of CCN in a CTM (page 2-7). In circumstances where other processes are relevant to changing aerosol particle size distributions—perhaps terms included in f_k, or other processes held constant in the model but changing in the actual atmosphere and therefore affecting the measured distribution—the result would presumably be a (not physically relevant) change to at least one of these investigated process rates. Is there a limit to how much the resulting error on the estimated process rate would be, or a method to identify when such a circumstance is occurring? Would it be evident in, say, the condition number of equation 3, or is there another method of detecting such a scenario and limiting its impact on process rate estimates?

This is of course an issue with all under-constrained inverse problems, not unique to this method, and is beyond the scope of this manuscript to resolve. However, some discussion of the issue may be warranted as an outstanding issue with such efforts.

2. Additional physical reasoning regarding the circumstances under which an ill-conditioned sensitivity matrix may occur.

The authors discuss a weakness of the method occurring when the sensitivity matrix, G_k, is ill conditioned (page 9-6). Cases in which solutions to these situations are discussed occurs at page 10-28 and 11-10. In these situations, it may be helpful to include more physical reasoning regarding the ill-conditioned scenario and the implications of removing one uncertain process rate (held constant for the given time step).

To a very general degree, additional discussion might address several open questions: Would this be interpreted as the aerosol process being generally sensitive to changes in more than one inventory variable? What are the physical implications of this in terms of adjustments to the aerosol process rates at such time steps? Would real world changes in other relevant processes, or those contained in f_k, be at least partially responsible? Moving forward with this method, should those be considered? How does this model perform when multiple process rates are changing simultaneously?

For example, in the ill-conditioned scenario described at the beginning of section 4, would the physical interpretation be that changes to both emissions and SOA production rates are expected by TOMAS to have roughly the same impact on N_10 and V_dry (similar sensitivities in G_k)—and that information on changes to the size distribution (via our inventory variables) are constrained to only influence emission rate to solve this issue? That would seem to be a reasonable solution when lacking a better reason to constrain one of the process rates, but more discussion of G_k and the condition number in such circumstances would be helpful. If this is not the correct interpretation of these ill-conditioned sensitivity scenarios, it may help to provide more physical reasoning of what is happening in the model and what the physical implications are in these circumstances.

Minor comments and technical corrections

1. When referring to the three investigated aerosol processes (nucleation, emis-

sions, and growth) throughout the manuscript, the order of these terms is occasionally changed (e.g. abstract line 1-14 "... aerosol nucleation, emissions, and growth rates..." and 1-19 "... aerosol emissions, growth, and nucleation..."). Readability may be improved simply by keeping a consistent order when referring to these processes.

2-14: The last several sentences in this paragraph are somewhat confusingly worded. Consider clarifying the meaning here.

3-1: The use of the word "integrates" could be misconstrued as integration of the size distribution here. Consider a different word such as combines or includes.

4-14: "as nearly as possible". Consider something like "as closely as possible".

4-19: The point of this sentence is valid, but these processes are not the only processes that control evolution of the particle size distribution.

6-2: The terms here look somewhat like t_k(mu_k) as a function. Consider a change to something like "...depend on the scaling factors (mu_k), at time t_k ..." if appropriate.

7-1: Should mu_k_hat be referred to as "scaling factors" or similar here, as in 6-25 and 7-11, to prevent confusion?

7-17: Even if restating from an earlier cited source, an additional reference for more information on the use of the condition number and relative sensitivity array in this methodology would be helpful for a reader wanting more information on this step.

7-24: Consider "e.g." rather than "i.e." if appropriate.

8-25: Refer to inventory variables for each of these in this sentence, i.e. "...underpredicted aerosol mass (via dry aerosol volume, V_dry) as well as N_10..." or similar.

9-5: What was the justification for these ranges of rates spanning expected atmospherically relevant process rates?

Section 3.1: Was the uncertainty and estimated effect of instrument noise calculated

using only the limited "meteorologically stable" time periods as opposed to the full year of data?

10-16: Should be "damps" rather than "dampens".

13-15: Use of "integrate" again here. Consider "... way to combine information from ..." or similar instead.

Fig 9: Needs a legend to show line color meaning in figure.

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