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Atmospheric Chemistry and Physics Discussions

Interactive comment on "MATRIX (Multiconfiguration Aerosol TRacker of mIXing state): an aerosol microphysical module for global atmospheric models" *by* S. E. Bauer et al.

Anonymous Referee #2

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This paper presents a new aerosol microphysics module for global models that incorporates a detailed representation of the aerosol mixing state. The mixing state representation in MATRIX is a significant advance over most current global aerosol models. The approach is similar to that of Jacobson (2001), but the non-sectional size representation makes it more efficient. The module is described in considerable detail. MATRIX was implemented in GISS ModelE, and representative results from two global simulations are provided. Results from limited box-model tests are also provided.

The paper focuses on description and some evaluation of the new module with limited new results, but this information is of interest, and more results will undoubtedly be

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provided in subsequent papers. The paper is certainly within the scope of ACP and should be of considerable interest to ACP readers. The paper is of good quality, and it should be published after referee comments are addressed.

Specific comments:

1. I concur with nearly all the comments made by J. Pierce.

2. Page 9932 (lines 4-5) and page 9965 (lines 15-17). MATRIX is based on QMOM. However, there is no mention at all of QMOM in section 2 or the appendices. The lognormal parameters in Table 1 and the use of the geometric standard deviation in Eqn. 18 lead to some confusion about what is actually happening in MATRIX. The 2-moment versions of QMOM and MIDAS have not been previously described, so some details about them and their application in MATRIX need to be included in the paper:

For MIDAS, is a single log-normal distribution used? Is the geometric standard deviation varied (as in Wright, 2000) or set to the values in Table 1?

What is the accuracy of the QMOM one-point quadrature for various processes (e.g., coagulation)? Also, what is the one quadrature point? E.g., is it the mass mean diameter?

Where in MATRIX are the QMOM quadrature and/or the MIDAS size distributions used for evaluating various process rates and integral aerosol properties?

What are the differences between the MATRIX approach (with its 2-moment QMOM and MIDAS) and a typical 2-moment modal approach that uses lognormals with prescribed geometric standard deviations? They appear to be very similar.

3. Section 1. Improved representation of the aerosol mixing state is a key feature of MATRIX. The introduction should provide a more detailed (although not necessarily exhaustive) review of previous work dealing with aerosol mixing state in global aerosol models.

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4. Page 9933 (lines 19-21). The modal method involves two assumptions: that the aerosol size distribution can be approximately represented by a set of modes, and that each mode can be approximately represented by a specified functional form (commonly lognormal). Moment-based methods do not require the assumption of modes, although this assumption is used in MATRIX.

5. Page 9933 (lines 24-28). This sentence, located immediately after the discussion of the sectional/modal/moment based approaches for aerosol modeling, gives the impression that the models listed all fall in the moment-based class. The Ackermann, Binkowski, Easter, Herzog, and Riemer models use the modal approach. Modal-based models all use moments in their algorithms, but they are distinguished from moment-based models by their assumption of lognormal (or similar) distributions for each mode.

6. Section 2 would be improved by adding more discussion of the rationale for and implications of the various assumptions made in MATRIX. For example, how were the 16 modes in aerosol mechanism 1 selected? Why was 5% chosen for the inorganics threshold that separates modes?

7. There is no mention of secondary organic aerosol in the paper. This is somewhat understandable, given that SOA is not explicitly treated in GISS ModelE. (Although, as noted in Pierce’s review, SOA from terpenes is included but is treated as primary organic aerosol.) Some limited discussion about how SOA might be incorporated in MATRIX would be of interest. In the current version of MATRIX, it would have been quite reasonable to distribute the OC from terpenes among several of the modes. Future versions of MATRIX will undoubtedly have explicit treatments of SOA.

8. Page 9935 (line 23). Please comment on the choice of organic carbon (OC) rather than organic matter (OM) as a model species, as using OC neglects some of the OM mass.

9. Page 9937 (lines 27-29). What is the reason for distinguishing BC-inorganics particles arising through condensation (BC2 and BC3) versus coagulation (BCS)?

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10. Page 9938 (lines 8-9). The MXX mode apparently includes both fine and coarse particles, so the size distribution of the MXX mode will be less accurately represented than for other modes. An alternative would be to have separate fine and coarse mixed modes. Please comment.

11. Page 9938 (lines 9-14). Why are the sea-salt modes treated differently (number not predicted) from the other modes? Also, why was this method for apportioning sulfate chosen? Was there a particular sulfate formation mechanism in mind?

12. Page 9943 (lines 6-10) and Section 5. The Napari et al. (2002) parameterization has been found to over-estimate nucleation rates [Anttila et al. (2005, Boreal Environment Research), Lucas and Akimoto (2006, Geophys. Res. Let.)]. Merikanto et al. (2006, J. Geophys. Res.) provide a newer parameterization of ternary nucleation This should be noted. The use of the Napari parameterization in the global model simulation is likely producing higher number concentrations in the Aitken mode than would be obtained using other parameterizations. This should also be noted, especially in the discussion of Figs. 15-16. The authors should consider repeating the simulations with a different nucleation parameterization.

13. Page 9947 (lines 9-14). This method for partitioning aqueous sulfate production among modes is equivalent to assuming that cloud drops formed on particles from different modes all have the same mean water volume. In reality, droplets that form on smaller particles (i.e., on CCN with higher critical supersaturations) tend to be smaller than those that form on larger particles. The partitioning approach used here (and in some other models) thus will tend to overestimate the growth rate of the Aitken-mode particles.

14. Page 9948 (top). What hygroscopicity is used for OC?

15. Page 9948 (lines 3-6). "Modes that are though of as insoluble cores with soluble shells, ... and bulk treatment of these parameterizations may underestimate this enhancement." Please clarify. I did not understand how this relates to what is being done

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in MATRIX.

16. Page 9950 (line 1). Cloud chemistry can also contribute to this growth.

17. Section 3.1. These tests do not use a highly accurate model (high-resolution sectional or high-order moment) as a benchmark, so the accuracy is not really known. It would be more appropriate to say that the use of the 0.5 h time-step causes only a small change in the results, rather than using terms like "quite accurate" and "very accurately predicted". Also, as there is no benchmark, these tests seem to be dealing with a fairly minor aspect of the accuracy of MATRIX.

18. Page 9954 (line 27) through 9955 (line 9). Please provide somewhat more detail about the removal processes. For the various removal processes (dry, in-cloud wet, below-cloud wet), are separate "first-order" removal rates calculated and applied for aerosol mass and number? Do the rate calculations use just the mean sizes of each mode or also the geometric standard deviations?

19. Section 5.2. Budget results for aerosol number, similar to Table 3 but with individual aerosol microphysical processes shown, would be of interest.

20. Fig. 12 and page 9961 (lines 1-14). It is surprising that dust mode contributions appear to be relatively small. Is most of the dust found in the MXX mode? Both here and in the discussion of Fig. 10, information on the composition of the MXX mode (both for boundary layer and lower free troposphere) would be of interest.

21. Page 9961 (line 15). TRACE-P focused on Asian outflow.

22. Figure 3. The greatly compressed vertical scales on these plots make it very difficult to compare the results from the different mechanisms.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 9931, 2008.