

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

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Received and published: 13 June 2008

This paper describes the incorporation of size-resolved aerosol microphysics into GISS ModelE. The aerosol microphysics is solved using a multi-population version of the aerosol method of moments (MATRIX). This is the first time that the aerosol method of moments has been used in a global atmospheric model and has the potential to determine aerosol size, composition and mixing properties efficiently relative to other techniques.

The authors begin by describing MATRIX. Several configurations of assumed aerosol populations are presented. Mathematical details are given on how the various aerosol

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microphysical processes affect the aerosol populations. Box model tests are used to test the accuracy of the method of moments to simulate some of the processes that I believe were not tested in the previous papers describing aerosol method of moments. Finally, simulations were ran in GISS ModelE and the results are compared to an array of observations.

The paper focuses on documenting and evaluating the new model. With the exception of the information contained about global aerosol mixing state in many of the final figures, the paper does not focus on presenting new findings; however, this is acceptable (if not desirable) because the paper is of suitable length and the initial documentation of the model is necessary. The paper is high quality, relevant to the ACP audience and deserves to be published in ACP once the suggested changes and clarifications below have been made to the text.

1. Page 9933, Lines 4 and 15: The citation Adams et al., 2001 is incorrect. This paper was before size-resolved aerosol microphysics was included. Please use “Adams, P. J., and Seinfeld, J. H. (2002). Predicting global aerosol size distributions in general circulation models, *J. Geophys. Res.* 107.”
2. Page 9935, Line 25 and Section 2.4.1: Its seems that water uptake from organic solute is ignored in the model. Obviously water uptake from organics is highly uncertain and greatly complicates the thermodynamic calculations; however, you should explicitly mention that you ignore it and that there may be underestimation of aerosol water.
3. Throughout paper, but mostly Section 2.3: “secondary particle formation” used to denote new particle formation (or aerosol nucleation). While this is by no means incorrect, it may be confusing to some readers who think of secondary aerosol as the condensation of mass onto particles but not necessary the creation of new particles (e.g. people who work with SOA). I suggest using “new particle

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- formation”; however, the reader should be able to determine your meaning from the context.
4. Page 9938, Line 13: Why are the number concentrations found from the mass concentrations by assuming a lognormal distribution and providing a geometric mean and sigma when you explicitly track the number concentration in the method of moments?
 5. Table 1: A continuation of the previous comment. How can both $D_{g,N}$ and $\sigma_{g,N}$ be specified for each population when you are tracking both number and mass of the population? You have three degrees of freedom for a lognormal distribution, but you have four parameters specified. It makes sense for the emissions, when you are given the mass of emissions from the inventory and you need to know the number and size of particles to emit, but not elsewhere in the model.
 6. Equation 9: What is the significance of the superscript “(3)” in on $f_i^{(3)}$?
 7. Page 9943, Line 16: The ionization rate varies not only with altitude, but also strongly with latitude (see Usoskin, I. G., and Kovaltsov, G. A. (2006). Cosmic ray induced ionization in the atmosphere: Full modeling and practical applications, J. Geophys. Res. 111.). Your assumption certainly doesn't need to be changed here, but it may be worth noting.
 8. Page 9946, Line 20: Is there a citation for the water uptake of sea-salt?
 9. Section 2.5: If I am correct, in the aqueous oxidation of sulfate scheme, the amount of new sulfate mass formed in each population is proportional to the number of particles activated in that population. This new sulfate mass is then added to the total mass of that population while the number is held constant. This means that all of the particles (both particles that were and were not activated) grow in mass because M/N has increased. Realistically, only the particles in the

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population that were activated should have grown. This is obviously difficult (impossible?) to account for with only 2 moments per population. If each population did not span a large size range then most populations would be either entirely activated or entirely not activated, but many of yours span somewhat large size ranges (relative to sectional models). It would be good to comment on the potential numerical diffusion that may occur here.

10. Page 9947, Line 19: Is the CCN activity of the various populations only determined from its solubility? If so, you should comment on how you assume that all solute is the same. For two completely soluble particles of the same size, the particle with a higher solute density will activate at lower supersaturation. For example, sodium chloride will activate at lower supersaturations than ammonium sulfate.
11. Page 9950, Line 17: Says “AK” instead of “AKK”.
12. Page 9951, Section 3: The description of the discrete model is vague (almost non-existent). Please elaborate. Does this model use the same population and moments as MATRIX, but numerically solve the aerosol general dynamic equation (rather than the analytical solutions given in the text)? Does it discretize the aerosol size distribution into many size sections rather than using the method of moments? The sentence starting at line 14 on page 9951 seems like it is supposed to give some insight into the discrete model, but I found it confusing. What do you mean by grid point here? Is this a size section or a moment or something else? Also in this sentence it says “than exist at point i”, but it should be “that exist at point i”.
13. Section 3.2: It doesn't seem as though the discrete model is used in Section 3.2. In the introduction to Section 3, the wording led me to believe that the discrete model would be used throughout the box model tests. Please clarify in the introduction to Section 3.

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14. Page 9954, Line 18: Terpene emissions*0.1 seems like a surrogate for SOA. With SOA, organic mass is added, but not number. It seems like you are emitting these as primary particles where you add both mass and number. If so, you should explain this. If not, you should probably refer to it as SOA.
15. Section 5.1: The section is titled “Mass simulation”. Are you only simulation mass here (i.e. ignoring the number moments)? Is “BASE” the same simulation that is discussed in Section 5.2 “Number and size simulation”? If the simulations are in fact different between these section, this needs to be explained. If the simulations are the same, it should be mentioned that the “BASE” simulation is used in Section 5.2 too. Also, if this is the case, it would be much more clear to name the sections something like “Aerosol mass sensitivity and comparison to observations” and “Comparison of aerosol number and size to observations”.
16. Page 9956, Line 18: It mentions that NO-MIC is lacking sulfate condensation processes. Does this mean that no mass is added through sulfate condensation? I would assume that you DO want condensation because bulk mass aerosol models certainly have condensation of gas phase sulfuric acid (they just don't have growth of aerosol size because the aren't predicting the sizes). Lack of condensation obviously would cause a big loss of sulfate. I do not know why you want to leave it out though if you are trying to represent a bulk mass model. This may be the only point where I may suggest redoing a simulation.
17. Page 9957, Line 25: “there is a large spread in mean diameter”. Does this mean that when you look at the mean diameters of all DS2 in all of the grid cells of the model, there is a large variation? Please clarify.
18. Page 9958, Line 7: “United Statues” :)
19. Figure 5 and associated text: Maybe try to quantify the average error and bias of the comparison and add this to the discussion in the text rather than just using

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- phrases like “good comparison to observations”.
20. Page 9958, last paragraph: There is discussion of the Costa Rica data, but not the Texas data (just the cloud scavenging stuff). I’m not sure if this was intentional.
 21. Page 9958, Lines 21-22 and Page 9959, Line 6: “Schwarz and et al (2006)” appears twice.
 22. Figures 5 and 6: Please explain what the dashed lines represent.
 23. Page 9959, Lines 15-17: Sizes were doubled... diameters were doubled or mass of particle was doubled? Also, was the total mass emitted held fixed (with the number of particles emitted reduced because of the size increase)?
 24. Page 9959, Line 26: “On a global averaged”
 25. Figures 10, 12 and 14 and associated text: In all of these figures, the modeled number is compared to the number of particles larger than 100-300 nm that were observed. Why is AKK included in this comparison when, I assume, most of its particles are smaller than 100 nm. For the instruments with a lower cutoff of 300 nm: I would assume that much of the accumulation mode number may be smaller than 300 nm, this should be removed from the comparison too.
 26. Figure 14 is not mentioned in the text.
 27. Figure 14 top panels: I’m really surprised that sulfate is virtually non-existent over the Eastern US. Is something wrong here? There seems to be plenty of inorganics in Figure 13. Why are nearly all of the particles in the BOC population that does not contain much sulfate? Is this because you actually are only looking at particles in the model larger than 300 nm? If so you can disregard my earlier comment about removing smaller particles from these comparisons, but it should be clarified in the text.

28. Figures 15 and 16: How come each population covers such a limited size range causing huge gaps in the overall distribution? Is this because of the pre-chosen sigma values? Will this go away when more moments per population are used?
29. Figures 15 and 16: This could be because the figures were shrunk to fit the ACPD half-pages, but the text is very hard to read. Please make all text bigger if possible.
30. Figure 15 and 16: Is it possible/desirable to smooth the modeled size distribution (blue line), so that its not as jumpy? This is probably related to the short size range comment above.
31. Page 9963, Line 14: “lag” should be “lack”
32. Page 9963, Line 25: “very successful simulation”, please be more quantitative with these assessments throughout.
33. Page 9964, Line 23: It is not clear how ignoring the kinetics of nitrate condensation would cause a lack of nitrate on the coarse mode particles. If anything, including the condensational kinetics may reduce the nitrate in the coarse mode because it takes the coarse mode much longer to reach equilibrium (see Meng, Z; Seinfeld, J.H. (1996). Time Scales to Achieve Atmospheric Gas-Aerosol Equilibrium for Volatile Species, Atmos. Environ. 30:2889-2900). It is possible that if nitrate is evaporating from particles that the large particles will hold on to their nitrate longer causing you to underpredict nitrate when you don't have condensational kinetics, but I would assume this would happen less often than the former case.
34. Page 9964, line 28: I think that condensation needs to be included in the NO-MIC model because bulk models have condensation.

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35. Page 9970, line 22: Peter Adams and I submitted a paper (in review with Aerosol Science and Technology) where we evaluated the pseudo-steady-state assumption for sulfuric acid. One of the things we tested is the error that you get in your particle number by assuming pseudo-steady-state even when the time scale for steady state is longer than the model timestep. The errors ended up being low. Often you nucleate earlier than you would have if you were explicitly tracking sulfuric acid vapor, but the nucleation is soon dampened by the increase in surface area, so the error is small. Obviously, your method is an improvement on this, but if you would like to discuss this in there I'd be happy to send a preprint. By no means will I be offended if you don't because this section is solid as is.

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