

Interactive comment on “A framework for expanding aqueous chemistry in the Community Multiscale Air Quality (CMAQ) model version 5.1” by Kathleen M. Fahey et al.

Anonymous Referee #1

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Kahey et al. replace the hard-coded aqueous chemistry mechanism in CMAQ with a flexible chemical mechanism using the KPP solver. This allows a more general approach, with aspects such as mass transfer and H⁺ concentration now allowed to evolve dynamically. The authors demonstrate this method by first replicating the existing AQCHEM mechanism (AQCHEM-KMT), and then extending it with a more complete representation of organic chemistry (AQCHEM-KMTI). This demonstration shows little impact on the model representation of sulfur chemistry, but secondary organic aerosol is significantly affected. Their approach sidesteps some issues of the previous sulfur-focused implementation, such as the application of inappropriately long timesteps to the calculation of rapidly-evolving SOA concentrations, and delivers a use-

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ful tool for other groups to explore other aspects of aqueous phase chemistry.

This paper represents an incremental development in the representation of aqueous phase chemistry in a regional-scale model. However, such a development, especially in a model as widely used as CMAQ, is both overdue and welcome. The reapplication of KPP to solve aqueous phase chemistry seems natural and well implemented, and I am pleased to recommend this paper for publication in GMD, pending some minor revisions. I have outlined two general criticisms below, followed by a list of minor recommendations.

My first and most significant issue relates to the question of computational efficiency. The final paragraph of the conclusions is informative, evenly discussing the computational trade-offs associated with the new, more complete aqueous chemistry mechanism. However, it references timing data which has not been given previously. Section 2.1 or 2.2 would benefit significantly from a dedicated discussion of the execution time of AQCHEM in each of the scenarios in comparison to AQCHEM-KMT. It would also be useful to know the degree to which some of the assumptions previously used by AQCHEM were found to be inaccurate. For example, with AQCHEM-KMT, the authors can diagnose the (in)accuracy of the electroneutrality assumption previously forced on the aerosol.

My second issue regards operator splitting. Previous work has shown that splitting gas- and aerosol-phase chemistry into two separate operations inevitably introduces errors, and that these errors can be quite large (Djouad and Michelangeli, 2004). This is especially pertinent now that mass transfer limitations are being explicitly considered. Unfortunately Kahey et al. do not state how their aqueous chemistry operator fits into CMAQ as a whole. My assumption is that it remains a separate operation from gas-phase chemistry, but without a clear explanation the reproducibility of the work is compromised. The paper would benefit from a description of the order of operation of AQCHEM-KMT with respect to the rest of the model operators.

Specific comments and technical corrections

The following minor issues should be addressed:

- Page 5, line 18: The sentence beginning “While in AQCHEM...” is confusing and would benefit from a rewrite.
- Page 8, line 13: Suggest that “Figures 2 b and d” is changed to “Figures 2b and 2d”
- Page 11, line 7: The phrasing “At least a couple micrograms per meter” is rather vague.
- Page 11, first paragraph: Some sense should be given of the relative impact of implementing AQCHEM-KMT. With only absolute differences given, it is difficult to tell if the changes are ever significant. Suggest the authors state what the maximum and average percentage change in the given grid cell is. Similarly, Figures 3 and 4 would benefit from an additional panel showing the “baseline” (AQCHEM) concentrations in January and July. Without this, it is difficult to tell the significance of AQCHEM-KMT’s changes.
- Figure 1a: The caption is unclear. Specifically, it should be stated exactly what is varying between different points for the same solver (ie that different tolerances are being tested, and over what range). It would also be helpful to point out explicitly what is changing between the different plateaus - it appears that SDA is more sensitive to relative tolerance than to absolute tolerance, based on the clustering behavior.
- All of the figures showing land area should have axis markers (latitude and longitude, if possible, or at least the X and Y dimension indices). Once these are in place, the grid index given on page 11, line 5 should be changed to match the

dimensions used for the figures, so that the reader can identify which point on the grid is being discussed.

- Table 2: There are some typesetting issues here, particularly where a subscript should or should not have been used. Note, for example, the “Other information” entry for wet deposition.

References

Djouad, R. and Michelangeli, D. V.: Investigation of splitting gas and aqueous operators in atmospheric multiphase box models, *Atmos. Res.*, 71(4), 253–263, 2004.

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