

We thank the reviewer for their helpful comments on the original version of the manuscript and respond to each point below.

Response to Anonymous Referee #2

This manuscript describes an implementation of a detailed chemistry scheme (MECCA) within a climate model (CAM). Three aspects are discussed: (1) alignment of chemical species between the host model and embedded chemistry scheme; (2) testing performance of three numerical integration methods available within MECCA; (3) balancing the computational load of MECCA within CAM for parallel execution. The work seems to have been performed carefully and is presented clearly. My main concern is that there are insufficient new developments for publication. Alignment of chemical species between a host model and an embedded chemistry scheme (that was not designed specifically for the host model) is a task undertaken by many model developers and, in the opinion of this reviewer, does not justify publication. Three Rosenbrock numerical integration methods (Ros-2, Ros-3 and RODAS3) were tested for computational efficiency and reproducibility of results. These solvers have been tested previously for a range of atmospheric chemistry problems (Sandu et al., 1997; Verwer et al., 1999) finding that all perform capably and that selection of one over another may depend upon the accuracy desired and the stiffness of the chemical problem. The results of this study are consistent with, but do not go beyond, results already published.

We consider this work to be novel on many fronts, and refer the reviewer to our response to reviewer one's question number 5 (see uploaded comments). We are aware of no studies that use KPP's Rosenbrock solvers in 0-d or 3-d coupled circulation/chemistry studies including global scale combinations of photo (cloudy and clear-sky), multiphase, mass-transfer, aqueous (both aerosol and cloud) and acid-base chemistry across the troposphere in the tested mechanism or any sub-mechanism of similar detail. Primarily, the inclusion of mass transfer and aqueous chemistry renders a far more stiff and unstable solution which was a serious challenge for the KPP solvers; and the impact of step-wise transients in the chemical and physical state of the simulated atmosphere as a result of 3-D advection further destabilizes the solution. Further, none of the studies we reviewed for this manuscript were explicit in their analysis with regard to chemical species whether in terms of their relevance to atmospheric chemistry or the difficulty in rendering them in fully coupled simulations. The fact that our results are consistent with others' results showing that the RODAS solver was a sufficient or superior option within KPP is indicative of the robustness of the Rosenbrock method. We point out that Verwer et al. 1999 only consider a small gas/photochemical mechanism, and that only in one scenario in Sandu et al. (1997) is aqueous chemistry considered. Neither of these mechanism, nor that used in other atmospheric chemistry models (e.g. see Henze et al., 2007) are sufficient to address the chemical complexity involving inorganic halogen cycling necessary to test global-scale hypotheses. A discussion of this result and our understanding of its context has now been included in the text. It may also be the case that our reliance upon supplemental for details of the reaction mechanism was insufficient; but we stress that the breadth of information in it requires that it be published as such. We therefore refer the referee to the supplement.

L215: "Other studies have investigated the stability and efficiency of the Rosenbrock solvers in KPP across a range of chemical scenarios (Henze et al., 2007; Verwer et al., 1999; Sandu et al., 1997). This study is the first we are aware of where KPP's Rosenbrock solvers were tested against such a complex chemical mechanism including gas, multiphase, and photochemistry through the entire atmosphere."

Figure 2 of the manuscript shows "the frequency distributions of average integration times (or waiting-time for completion of one chemistry timestep) for all grid cells varied among the solvers tested" and is the most interesting result presented. The distributions for Ros-3 and Ros-2 tail toward long integration times and there is discussion that this tail interferes with the load-balancing scheme and slows down

model execution. The authors propose that improved load-balancing schemes could be developed by recognizing characteristics of the distribution of chemistry integration times. Such schemes would be valuable to the modeling community and their inclusion would improve this manuscript and merit publication.

We agree that such a load-balancing scheme would be valuable to the modeling community but the development of such a scheme would correspond to a substantial research effort that is well beyond the scope that could be supported through this project. Current efforts, as a separate project, involving both the system described here and Harvard University's GEOS-Chem atmospheric chemistry CTM, are in progress that will address these questions.

Sandu, A.; Verwer, J.G.; Blom, J.G.; Spee, E.J.; Carmichael, G.R.; Potra, F.A. 1997. Benchmarking stiff ode solvers for atmospheric chemistry problems II: Rosenbrock solvers. Atmospheric Environment, 31:3459-3472. Doi:10.1016/S1352-2310(97)83212-8

Verwer, J.G.; Spee, E.J.; Blom, J.G.; Hundsdorfer, W. 1999. A Second-Order Rosenbrock Method Applied to Photochemical Dispersion Problems. SIAM Journal on Scientific Computing, 20:1456-1480. Doi:10.1137/S1064827597326651