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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A computationally-efficient secondary organic aerosol module for three-dimensional air quality models" *by* P. Liu and Y. Zhang

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This paper presents an analysis of how CPU time can be reduced by relaxing stringency criteria for an aerosol model, along with a statistical fitting of the UNIFAC approach. Such an exercise is valuable, and the CPU speed-up obtained with a parameterized UNIFAC was interesting, but this paper has however a number of major shortcomings if the goal is to provide techniques for use in 3D atmospheric models.

Most importantly, the test cases considered are too few and too simple. A simple box model is used, with no emissions, no deposition, and a very long (24h) integration period. This is not an appropriate way of testing solvers designed for 3-D models, in which the chemistry is essentially re-started after each advection time step (e.g. 5-20

mins). One long simulation leads to a rather gentle development of the chemical processes, and highly favours some types of solvers (especially Gear-type) over others which are better suited to rapid-restarts. The paper by Saylor and Ford (AE, 1995) stressed this problem, and suggest approaches to provide tests which are more appropriate for 3D models. Another major problem of this paper is the very restricted number of test cases used, only four, and their lack of consideration of non-typical conditions. Real 3D modeling exposes the numerical scheme to a huge number of different concentration regimes, and it is important to try to cover the extremes of these regimes as well as more typical situations. It is the extremes which result in numerical instability or convergence problems. A numerical scheme which works fine for the "typical sunny day" as discussed in this paper may well run into severe trouble for less typical situations, for example an urban plume moving into a forested area in the middle of a heat-wave. For example, Hesstvedt explored 21 scenarios, with a factor 100 range in both NOx and HC levels, and found very different accuracy for different cases.

In this context, the maximum isoprene concentration of 2 ppb is very low and certainly not sufficient to constrain a numerical scheme. Even for regional modelling in the U.S. much higher isoprene and BVOC levels need to be considered. For example, de Gouw et al (JGR, 2005) estimated isoprene concentrations in OA source areas to be of order 10 ppb. The recent review of Heald et al. (ACP, 2008) found daytime mean total observed organic carbon concentrations of between 4 to 456 ug/m3 over North America - 3D models need to cope with this full range!

Other conditions which need to be addressed in a 3D models include those associated with the free troposphere, where most concentration levels are low, but where low temperatures encourage condensation of SOA - conditions far removed from those presented in this paper.

As a final example, global models need to deal with SOA formation over for example the Amazon, where NOx concentrations are less than 100 ppt but isoprene can easily be several ppb.

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p7093, line 5. It is stated that the gas-phase chemistry requires 2.1s CPU versus 199.3s for the aerosol phase. This sounds like a very ineffective aerosol-phase solver to me, so the authors might be better advised to look for very different methods rather than trying to relax stringency in search of CPU savings. UNIFAC is a rather extreme choice for application within an aerosol dynamics model, and the authors should at least discuss alternative possibilities.

Also, if the gas-phase consumes so little CPU, why bother presenting results from QSSA and SMVGEAR? Aren't they irrelevant? What is the CPU split between partitioning and UNIFAC, with different test cases? This is crucial information which I didn't find.

The presentation of results is also rather unsatisfactory. The type of accuracy vs CPU time plots shown e.g. in Sandu et al. (1996) provide very valuable information on the behaviour of different solvers. Here the authors present just a few sentences on accuracy loss, and one figure showing average percent deviations. (And this figure has a caption which isn't explained - what do the different symbols mean? Readers should not need to search the text to find out.)

Most statements are made of the form, "reduction in some strigency requirement by X% reduces CPU by Y%". Very little information is presented on the actual costs in terms of accuracy, except some rather vague sentences. It isn't even clear if the authors are citing average percentage deviations, or peak errors. In many cases, the accuracy obtained sounds unacceptable to me. If a scheme gets major species wrong by 5%, presumably it gets some of the "minor" species wrong by very significant margins. These minor species are often of importance for air pollution modelling.

Other papers dealing with numerical methods tend to aim at something like 1% accuracy.

p7092, Line 22 I am puzzled by the poor performance of the QSSA and Rosenbrock schemes when they are run with most stringent error-tolerance, with 4.2% for ROS3

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and 5.6% for QSSA. This suggests to me a poor setup of these solvers, since they should normally perform very well when short time steps are used. (This paper doesn't say what timesteps were used for these solvers - this is rather crucial information). For example, Sandu et al. (1996) found SDA values of 4 (0.01% accuracy!) for ROS3 at the most stringent settings. Hesstvedt et al (1978) found maximum deviations for ozone of -1.76% for a 30 second time step were reduced to 0.23% for a 5 second timestep for QSSA. Further reductions in time-step would have given even better results.

All in all, I was convinced that relaxing stringency reduced CPU, but I had little idea of whether the scheme was usable in real atmospheric conditions.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 7085, 2008.