

Interactive comment on “Modeling natural emissions in the Community Multiscale Air Quality (CMAQ) model – Part 2: Modifications for simulating natural emissions” by S. F. Mueller et al.

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The first part of the comment requests information on the effect of anthropogenic emissions on the model response to the introduced organic sulfur chemistry. The overall effect on atmospheric sulfur dioxide and sulfate is illustrated in Fig. 1 which plots the change in SO₂ and sulfate relative to a model simulation based on the original sulfur chemistry. Large relative increases in both species occur over the oceans because of the organic sulfur chemistry and the very low levels that occur in those locations in the absence of such chemistry. Inland over the eastern United States (where most

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anthropogenic emissions occur) the changes are much smaller and are near zero (or smaller than ± 25 percent) in most model grid cells. In the east there are areas of both increases and decreases of SO₂ that tend to cancel out. Sulfate is more likely to increase rather than decrease but this is due in large part to sulfate advection from off the oceans and cloud chemistry model changes that tend to increase sulfate even in the absence of organic sulfur. No systematic large sulfate changes are seen that can be clearly traced to man-made SO₂ emissions (for example, in the Ohio River valley) although this does not mean short-term effects do not occur that do not appear in the plot. Note that this topic is further addressed in a third manuscript that describes results from modeling both natural and full emission scenarios using the revised CMAQ model.

It is true that the aerosol module in CMAQ does not allow interactions between the coarse and fine particle modes. The primary reason for this is that there is little evidence that significant sulfate exists in the coarse size mode. We believe the commentator is referring to acid catalysis of organic material leading to secondary organic aerosol (SOA) formation. Coarse particles tend to be alkaline (soil dust containing calcium or sea salt particles) and it requires a great deal of acid particle migration to the coarse mode to enable acid catalysis. In a world of natural-only emissions there is even less acidity to migrate to the coarse size fraction and this would make it even less likely that enhanced SOA formation could occur. Given the unlikelihood of significant coarse particle acidity we believe the potential effects on SOA from mixing particle modes are extremely small.

A third comment is on the influence of background ozone on model results. We were able to make our comment about ozone background because of other information provided by the model that was not included in this paper. The third manuscript, alluded to previously, contains a detailed examination of the influence of background concentrations on simulated ozone. These results demonstrate clearly that it is the background information provided by GEOS-Chem that determines much of the grid-average ozone

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behavior that we describe. The commentator appears to be using the term "background conditions" to also refer to initial model conditions. The initial conditions of all our simulations were not based on GEOS-Chem but instead on an assumption of "clean" air and all pollutant levels were initialized at very low levels. It is true that GEOS-Chem boundary conditions include anthropogenic influences but given the location of the boundaries those emissions are from northern Canada, southern Mexico, and Asia. Hence, North American man-made emissions have an extremely small influence on pollutants advected into the domain based by way of GEOS-Chem data. Those data were averaged for one month periods to define CMAQ boundary conditions so any annual cycle in global pollutant transport should be included. Thus, grid-averaged changes in ozone are in response to changes in boundary conditions relatively unaffected by North American emissions but strongly influenced by pollutants from farther upstream in the atmosphere. The reviewer is invited to view the third manuscript after it is posted as a discussion paper to learn more about this issue.

We appreciate the fact that this commentator identified several potential editorial problems with the paper and we will address these when we prepare the final manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 15811, 2010.

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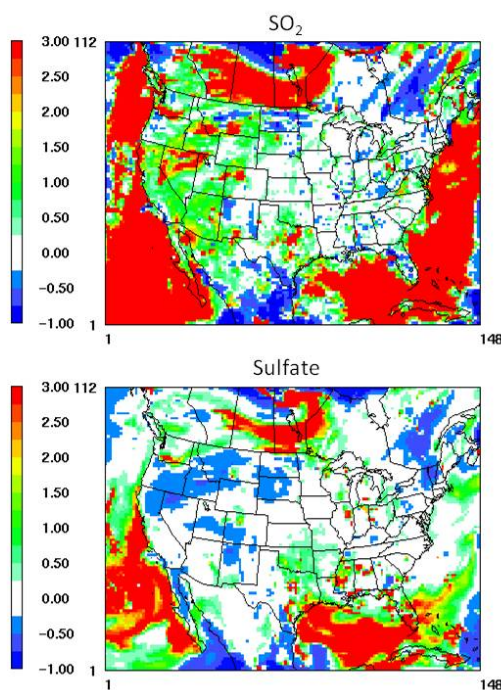


Fig. 1. Fig. 1. Relative changes in surface sulfur dioxide (top) and sulfate aerosol (bottom) in July due to the introduction of new sulfur reactions into the CMAQ model.

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