Geosci. Model Dev. Discuss., 2, C571–C576, 2010
www.geosci-model-dev-discuss.net/2/C571/2010/

Geoscientific
Model Development
Discussions

Interactive comment on "Simulating emission and chemical evolution of coarse sea-salt particles in the Community Multiscale Air Quality (CMAQ) model" *by* J. T. Kelly et al.

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Received and published: 27 February 2010

We thank Referee#2 for constructive comments and a positive assessment of our study. These comments have reaffirmed our statements in the manuscript about the need for continued development of sea-salt emission parameterizations applicable to diverse coastal conditions. Below, we have copied the referee comments in italics and inserted our responses in standard font where appropriate. Note that the line and page numbers in our responses refer to those of the article published in GMDD.

The manuscript presents the incorporation of a sea-salt emission module and the improvement of the coarse particle mode in the CMAQ. The authors then apply the model

to a special study of a coast site in Tampa, Florida 2002 and show improve predictions of aerosol species, in particular nitrate as comparing to the previous version of CMAQ. The introduction provides a good review of various aspects in examining sea-salt particle. In modeling section, the authors clearly described the development of the module which is scientifically sound. The paper is well written and useful, I would recommend for potential publication.

My major concern is sea-salt emission module which developed usually base on data collected at limit surf zone that may not necessary apply to other areas.

In several places in the manuscript (e.g., p., 1345, line 28; p. 1346, lines 1-8 and 15-24; p. 1350, lines 19-22; p. 1356, lines 25-27; p. 1357, lines 1-7; p. 1358, lines 5-6), we mention the limitations of typical emission parameterizations for sea salt from the coastal surf zone and the need for a generalized parameterization that reliably adapts to local conditions. In the absence of such a parameterization and the necessary information on coastal features (e.g., bathymetry) in all regions, our simple approach is reasonable. This approach has improved CMAQ model performance for conditions of the BRACE study as well as for summer and winter conditions at nine coastal CASTNET and four coastal SEARCH sites in the eastern U.S. (Foley et al., 2009). In Fig. 1 and Section 2.2., we compare our sea-salt emission flux function with those derived from surf-zone measurements in California and Hawaii to understand how our emission flux might differ from those measured in other regions. This comparison demonstrates decent agreement among the flux functions, although the flux at high wind speeds is significantly larger for the California-based flux function. The comparison suggests that our flux function may be considered conservative in the sense that it produces somewhat lower emission fluxes than the other functions. As mentioned in the manuscript in Section 2.2, we used the California-based flux function of de Leeuw et al. (2000) in our preliminary model testing, but we decided not to include it in the CMAQ release because it produced large over-predictions of sodium at the Azalea Park site. While we acknowledge here and in the manuscript that our parameterization may not be ideal for all coastal conditions, our model performance evaluations indicate that it is an improvement over the earlier approach, which neglected altogether the enhanced emission of sea-salt particles from the coastal surf zone. As mentioned in Section 2.2, we plan to revisit this topic in the future as new approaches become more established. Also, as mentioned below, our model developments are currently being evaluated for locations such as Texas, California, Hong Kong, and Vancouver, BC.

This may have some implication to the CMAQ in regulatory applications to set the PM standard.

The implications of our model developments are likely to be small for regulatory modeling applications in the U.S. for several reasons. First, the U.S. EPA's National Ambient Air Quality Standards specify limits on observed concentrations of fine particles (U.S. EPA, 1997, 2006), whereas our model updates mainly impact predictions for coarse particles. For instance, in Section 4.2, we report that "the mass of ammonium in the coarse mode is on average only 3% of that in the fine modes, and so uptake of ammonia by the coarse mode does not significantly impact the fine particle distribution." Second, the drop in sodium concentration from the coastal Azalea Park to the slightly-inland Sydney site indicates that the impact of our updates will generally be minor outside of coastal regions. Third, in regulatory modeling in the U.S., the ratio of results from reduced-emission and base-case-emission scenarios is used rather than absolute pollutant concentrations (U.S. EPA, 2007). The use of such ratios (i.e., relative response factors) would dampen the impact of our model updates in regulatory modeling applications. However, our model developments provide a more realistic representation of air pollution processes and improve model performance, and so these updates have positive, if small, implications for regulatory modeling applications.

I would suggest a discussion about the sensitivity of the sea-salt emission module on different meteorological conditions and regions of the ocean. A test in the far inland monitor with less sea-salt emission influence would be help to tell the contribution of the increase prediction of nitrate due to sea-salt emission or aerosol module.

For the open ocean, CMAQ's parameterization of sea-salt emissions is based on the Gong (2003) flux function and the white-cap method, which were developed and tested elsewhere. References to studies describing this approach and its development are given in Section 2.2. Since this method is used in many atmospheric models and earlier versions of CMAQ, an extended discussion of the topic is not warranted in our study. For the coastal surf zone, we compare our sea-salt emission flux function at two wind speeds with those based on measurements in California and Hawaii in Section 2.2. Source functions for sea-spray particles produced over the surf are also compared in Fig. 8 of de Leeuw et al. (2000). These comparisons suggest that our approach is reasonable given current knowledge in this area. In addition to evaluating predictions in the eastern U.S. (Foley et al., 2009) and at the BRACE sites (this manuscript), we are currently involved in studies that evaluate CMAQ sea-salt predictions in Houston, California, Hong Kong, and Vancouver, BC. The combined knowledge gained from all of these studies will provide guidance for our future model developments. For inland sites, a preliminary evaluation of CMAQv4.7 predictions of coarse nitrate has been conducted (Bhave and Appel, 2009). This evaluation indicates that our model developments improve coarse-particle nitrate predictions at inland sites compared to standard CMAQv4.6, which does not simulate coarse-particle nitrate formation. However, at inland sites, coarse-particle nitrate formation due to reactions with carbonates in mineral dust (e.g., Kelly and Wexler, 2005) may also be significant. Therefore errors in coarse-particle nitrate predictions at inland sites may not be directly attributable to CMAQ's sea-salt emission parameterization. Reaction pathways for nitrate formation in dust particles are not yet included in CMAQ.

How much the new release will affect the fine mode particle and ozone predictions?

The new release (CMAQv4.7) incorporates many updates in addition to those described in our study. Foley et al. (2009) provide an overview of these model developments and evaluate their impact on fine-particle and ozone concentrations. We clarified this point in Section 2.4 of the updated manuscript. As mentioned in Section 4.2

and illustrated in Fig. 5, our model updates had a minor impact on fine-particle concentrations for conditions of the BRACE study. The impact of the updates described here on ozone predictions and on fine-particle concentrations away from coastal regions is negligible.

The regulatory application modeling usually set at 12 km grid, does the new release require higher resolution for sea-salt emission calculation?

The grid resolution used in regulatory applications in the U.S. is often finer than 12 km. For example, regulatory modeling in California is based on 4-km horizontal grid resolution. The 8-km resolution used in the current study is applicable to regulatory modeling applications. However, the sea-salt emission parameterization does not require especially fine grid resolution to improve model performance compared with CMAQv4.6. For our 32-km outer-domain simulations, model performance was better for the updated models (i.e., CMAQv4.6b and CMAQv4.6c) than for standard CMAQv4.6. Also, as mentioned in Section 5, Foley et al. (2009) report improved performance associated with our model developments at nine coastal CASTNET and four coastal SEARCH sites for simulations based on 12-km horizontal resolution. While the optimal grid resolution varies depending on the application, our model updates improve CMAQ model performance for grid resolutions typical of regulatory modeling applications. In conjunction with CMAQ model releases, we provide a tool for converting data from GIS-based shapefiles into the gridded format (i.e., OCEANfile) required for CMAQ simulations. This tool, which is described in Section 2.2 of the revised manuscript, can be used to obtain sea-salt emission estimates at any grid resolution.

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