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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.

J. T. Kelly et al.

jkelly@arb.ca.gov

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We thank Referee#1 for constructive comments and a positive assessment of our study. The manuscript has been strengthened by revisions made in response to the referee's comments. In particular, the time-series plots for HNO_3 and HCl that have been added to the manuscript supplement provide additional evidence of the soundness of our model development. 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.

In this manuscript the authors present the sea-salt emission module, as well as an

improved coarse particle mode, that they have incorporated in CMAQ. They then applied the coupled MM5-CMAQ modeling system in the Tampa, Florida, USA area to evaluate the performance of the new version of the code with respect to the old one. The paper is well written and clear and potentially publishable. The modules that the authors have developed are scientifically sound. In addition the authors clearly present the assumptions and the limitations of the developed modules.

My main concern is in the application of the model and the associated attempt to explain the discrepancies between predictions and observations. Throughout section 4 the authors compare predicted and observed aerosol concentrations and speciation. They forget to mention all the other relevant processes completely.

We disagree with this comment. The possible influence on our results of processes such as deposition and transport as well as model grid resolution is mentioned in the Abstract, Section 4, and the Closing remarks: e.g., p. 1336, lines 26-27; p. 1349, lines 8-9 and 23; p. 1350, lines 15-23; p. 1352, lines 18-19 and 22-24; p. 1353, lines 1-3; p. 1355, lines 14-15; and p. 1357, lines 8-10.

Of main importance in predicting aerosol concentrations is, in addition to having an accurate aerosol module, to have correct transport, chemistry and deposition modules. The question arises on whether the differences between predictions and observations are due to inaccurate sea-salt emissions and deposition, or other modules in CMAQ need improvement.

The purpose of this study is to evaluate updates to CMAQ's sea-salt emission parameterization and the introduction of a dynamically interactive coarse-particle mode. To this end, we conducted simulations with standard CMAQv4.6 and two non-standard versions of CMAQ, which are identical to CMAQv4.6 except that they include one or both of the new model features. The updated models performed better than standard CMAQv4.6 in our study, and the better performance is clearly attributable to our model developments. Specifically, the improved sodium and chloride predictions are primarily due to the introduction of a parameterization of sea-salt emissions from the coastal surf zone, and the improved nitrate and nitrate-partitioning predictions are primarily due to the introduction of a dynamically interactive coarse-particle mode. Therefore we conclude that our model developments are sound. While other aspects of CMAQ may certainly require development, our study is focused on specific model developments rather than being an overall evaluation of the CMAQ modeling system. Broad evaluations of CMAQ are available elsewhere (e.g., Appel et al., 2007, 2008; Eder et al., 2009, Foley et al., 2009). Therefore we did not pursue additional analyses in response to this comment. For an overall evaluation of the CMAQ modeling system, we refer readers to the article by Foley et al. (2009) on pages 1341 (line 3) and 1358 (line 2).

What is the role of aerosol precursor chemistry in the gas-phase? Is the total HNO3 concentration predicted correctly in absolute values (ppb)? Are ozone, SO2, NOx predicted accurately during the testing period? Figures containing observed vs predicted concentrations of at least some of the above gases will give a feeling on whether chemistry and transport of pollutants is treated adequately by CMAQ.

The role of precursor chemistry is different for different particle components. Although we examine predictions of five inorganic particle components in this study, the two most relevant components for testing our model developments are sodium and nitrate. Sodium is a key component for evaluating our simulation of sea-salt emissions from the coastal surf zone, and nitrate is a key component for evaluating our simulation of chloride displacement from sea-salt particles. Sodium is non-volatile and non-reactive in the model, and so sodium predictions are essentially independent of model performance for gas-phase species (i.e., there are no gas-phase precursors for sodium). Therefore our evaluation of the sea-salt emission parameterization does not warrant a detailed evaluation of gas-phase chemistry. The gas-phase precursor of particle nitrate in the model is nitric acid. Since our model development includes a new treatment of mass transfer of nitrate between coarse particles and the gas phase, we conducted a detailed evaluation of CMAQ's nitrate-partitioning predictions in Section 4.3. This

evaluation demonstrates that our new model features greatly improve predictions of the gas-particle partitioning of nitrate. Therefore our mass transfer algorithm should reliably distribute nitrate (and chloride) between the particle and gas phases even in situations where absolute concentrations of the components disagree with observations.

In response to this comment, we added time-series plots to the revised-manuscript supplement that compare modeled and observed concentrations of nitric and hydrochloric acid at hourly time resolution during the study period. These plots are cited in Section 4.3 of the revised manuscript and indicate that our model updates greatly improve predictions of absolute concentrations of HNO_3 and HCl. Ozone, sulfur dioxide, and nitrogen oxides are of relatively minor importance to the model developments in our study. However, the good model predictions of particle sulfate and total nitrate suggests that gas-phase chemistry related to ozone formation and the oxidation of sulfur dioxide and nitrogen oxides are well represented. Also, note that the SAPRC99 gas-phase chemical mechanism used here has been extensively evaluated elsewhere (e.g., see http://www.engr.ucr.edu/~carter/) and is considered a state-of-the-science mechanism. Therefore a detailed evaluation of this mechanism is not warranted in our study.

Otherwise either chemistry or transport or both may be partially responsible to the poor performance of the model in reproducing inorganic aerosol concentrations. Of course in the latter case MM5 results should be examined to see whether the correct windfields are reproduced by the mesoscale model.

We are not sure what "poor performance" the reviewer is referring to. The performance of the updated model is generally quite good in this study. Perhaps we could have placed more emphasis on the good model performance and focused less on the limitations, but we were trying to highlight areas for future model development. One aspect of model performance that requires future study is the increasing low bias in sodium concentration with increasing distance from the Gulf of Mexico. While this trend could be due in part to errors in the wind fields, there is some evidence to suggest that this is not the case. Nolte et al. (2008) conducted a CMAQ-UCD model simulation based on the same MM5 output used here and did not find a low-bias trend in sodium predictions. Since predicted peak-concentration diameters agreed with observations and predicted particle distributions were narrow in that study, we indicate in the Abstract and Section 4 that the low-bias trend in our results could be due to too-rapid particle dry deposition caused by over-predictions in particle mode diameters and geometric standard deviations. In the revised manuscript, we also include this point in Section 5. A follow-up study is currently underway to investigate this question in more detail by comparing results from commensurable versions of CMAQ and CMAQ-UCD.

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