

## ***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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We thank Referee#4 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, these comments led us to create a much clearer version of Fig. 2 and to clarify our discussion of representing non-sodium sea-salt cations by equivalent concentrations of sodium. 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.

C583

*This study examines incremental changes to parameterizations of sea-salt emissions from the coastal surf zone and the dynamic transfer of HNO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>, HCl, and NH<sub>3</sub> between coarse particles and the gas phase in CMAQ. It fits well within the scope of GMD and provides useful information on the treatment of sea-salt particles in CMAQ. The manuscript is overall well written, clear, straightforward, and convincing.*

*It would probably be an exaggeration to say that new general insight into sea-salt treatment in advanced modelling systems (such as CMAQ) has been gained here.*

We do not claim that our study provides the type of insight suggested by the referee. However, our study appears to represent the first time CMAQ predictions of speciated particle size distributions have been evaluated with detailed (i.e., MOUDI) observations. Also, our development and testing of a new parameterization for sea-salt emissions from the coastal surf zone will be useful and of interest to modelers who focus on air quality in coastal regions.

*This study provides a critical perspective on current knowledge on the subject and on the methods used to investigate it. The modelling approach has been thought through and is sound, though it somehow assumes that discrepancies in the modelling of inorganic aerosol species stem from the treatment of sea-salt particles.*

We do not assume that model results for inorganic aerosol species stem from the treatment of sea-salt particles. For instance, sulfate and ammonium concentrations are not heavily influenced by the treatment of sea-salt particles. The main source of particle sulfate in the simulations is sulfuric acid condensation on fine particles, which have large surface area. Sulfuric acid is non-volatile in the model and condenses on any existing particle surface or nucleates to form new particles, and so sulfate concentrations are not strongly dependent on sea-salt concentrations. However, a small amount of primary sulfate is emitted in sea-salt particles, and so sea-salt emissions do have a minor impact on sulfate concentrations (as demonstrated by our model inter-comparison).

C584

The main source of particle ammonium in the simulations is ammonia condensation on fine particles that have been acidified following sulfuric acid condensation and dissociation. Ammonia condensation is generally favorable under these conditions because the vapor pressure of ammonia is inversely proportional to the hydrogen ion concentration in the particles. Our similar modeled ammonium concentrations for simulations with and without enhanced sea-salt emissions from the coastal surf zone demonstrate that ammonium concentrations are not strongly impacted by sea-salt concentrations. Of course, our finding that sodium concentrations are strongly dependent on sea-salt emissions is reasonable, because oceanic emissions are the only source of sodium in the model, and sodium is modeled as non-volatile and non-reactive. As for nitrate, we did not assume that its concentration is strongly dependent on sea-salt concentration; instead, we demonstrated this by evaluating the partitioning, absolute concentration, and normalized concentration of nitrate and by comparing results of CMAQv4.6 with those of CMAQ versions created to isolate such effects. Similar analyses were used to establish the relationship of chloride with sea-salt and nitrate.

*I am pleased to recommend the manuscript for publication in GMD after the authors clarify a few minor points (some editorial), which are detailed in the following. Counting all the lines and referring to the page number:*

*P1339, L10: Please correct 'disposition' to 'deposition'.*

This sentence is reworded in the revised manuscript to more clearly indicate the objectives of the BRACE study.

*P1345, L17-19: The sentence 'Those cation concentrations are downscaled during post-processing of the model output for comparison with observed sodium concentrations.' needs to be clarified. By the way, should 'Those cation concentrations are' read 'The concentration of those cations is'? Please explain how the downscaling of the concentration of cations is done with respect to their emissions.*

C585

Good point. We clarified this issue in the revised manuscript by inserting the following paragraph:

In CMAQ, primary sea-salt particles are speciated into three components (weight % by dry mass):  $\text{Na}^+$  (38.56%),  $\text{Cl}^-$  (53.89%), and  $\text{SO}_4^{2-}$  (7.55%). This speciation represents non-sodium sea-salt cations (e.g.,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ , and  $\text{K}^+$ ) by equivalent concentrations of sodium (on a molar basis) to achieve electroneutrality with the  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  anions. Moya et al. (2001) demonstrate that this approach is a good approximation when using thermodynamic aerosol models that do not include all crustal elements (e.g., see Fig. 2 of Moya et al, 2001). To recover the sodium fraction of sea-salt cations for comparison with observations, the modeled sodium concentration (i.e., sodium plus non-sodium sea-salt cations) is multiplied by a factor of 0.78 during post-processing.

*P1346, L23: I would suggest replacing 'is reasonable' by 'sounds reasonable'.*

This is a minor point, and we chose not to edit the manuscript here.

*P1348, L15: Please change 'on 15 days' to 'for 15 days' and qualify 'collected' on L14 to specify the frequency of the sampling (e.g. 'collected daily?', 'collected continuously'?).*

Good point. We clarified this issue in the revised manuscript by stating that samples were collected during 23-h periods on the dates specified in the supplementary figures.

*P1349, L25: Should 'CMAQv4.6' read 'CMAQv4.6b'?*

Actually, the statement holds true for both CMAQv4.6 and CMAQv4.6b, and we originally intended 'CMAQv4.6'. However, since this paragraph focuses on comparing CMAQv4.6c with CMAQv4.6b, we use 'CMAQv4.6b' in the revised manuscript to avoid confusion.

*P1350, L13: Please add '(Fig. 3)' after 'improvement'.*

This sentence was reworded in response to Referee#3's comments and now refers to Table S1.

C586

*P1352, L3: I would replace 'biggest' by 'largest'.*

Done.

*P1357, L12-13: Please change 'though, because measurements are not available' by 'as measurements were not available'.*

This sentence is reworded in the revised manuscript.

*P1357, L25: I would delete '; mean|. . . |N'. C and N are not defined and the notation is different from that used in the footnotes of Table 2 on P1367.*

Done.

*P1367, footnotes of Table 2: C and n are not defined and the notation is different from that used on P1357, L25.*

C and n are defined in the revised manuscript.

*P1369, Fig. 2: Please modify the figure so that the land/sea contrast is visible.*

Good point. We created a much nicer figure showing the inner modeling domain with a window for the Tampa region. Color coding is used to accentuate the land/sea contrast.

*P1370, caption of Fig. 3: Please change 'dashed line represents 1:1 ratio' to 'the dashed line represents the 1:1 ratio'.*

Done.

*P1371, caption of Fig. 4: Please change 'dashed line represents 1:1 ratio' to 'the dashed line represents the 1:1 ratio'.*

Done.

*P1373, caption of Fig. 6: Please change 'Tick marks represent' to 'The tick marks'.*

This sentence is reworded in the revised manuscript.

C587

Reference:

Moya, M., Ansari, A. S., and Pandis, S. N.: Partitioning of nitrate and ammonium between the gas and particulate phases during the 1997 IMADA-AVER study in Mexico City, Atmos. Environ., 35, 1791-1804, 2001.

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Interactive comment on Geosci. Model Dev. Discuss., 2, 1335, 2009.

C588