

Response to Anonymous Referee #1 comments on “Updating sea spray aerosol emissions in the Community Multiscale Air Quality (CMAQ) model version 5.0.2”

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This paper presents results of a model-measurement comparison that was done in order to improve sea spray aerosol emissions in coastal and near-coastal regions. It is a valuable paper in that measurements were used to improve model output. It should be publishable in GMD once the concerns below have been addressed.

We appreciate the reviewer’s comments and have responded in bold typeset to the individual comments below.

The title and abstract should state that the paper focuses on updating SSA emissions in coastal regions.

We’ve adjusted the abstract in the updated manuscript to reflect the fact that most of the model evaluation is focused on coastal sites and that these changes will have the biggest impact on coastal areas.

Throughout – use Revised and Baseline in text and figure captions to describe v5.0.2h vs. v5.0.2a. That will make it much easier for the reader to track which model version is being referred to.

We’ve renamed the simulations to this suggested nomenclature throughout the updated manuscript.

p. 3907, line 3: The Pierce and Adams (2006) paper estimates emissions of sea salt using a global model. Papers that report the sea salt fraction of CCN based on measurements should also be cited here.

The updated manuscript now includes the following: “Sea spray aerosols (SSA) contribute significantly to the global aerosol burden, both in terms of mass (Lewis and Schwartz, 2004) and cloud condensation nuclei concentration (Murphy et al., 1998; Pierce and Adams, 2006; Clarke et al., 2006; Blot et al., 2013).”

p. 3911, Lines 9 – 17: Were all measurements (and, therefore, cut-off diameters) at ambient RH?

The updated manuscript states that all size-resolved measurements were taken under ambient RH.

p. 3911, last paragraph: Why were the shipboard measurements made during CalNex not included in the analysis?

The updated manuscript now includes the following: “Although the CalNex campaign also included ship-based measurements of aerosol composition in conjunction with the Sea Sweep (Bates et al., 2012; Crisp et al., 2014), the portion of the cruise that took place in

June 2010 was mainly in the vicinity of San Francisco Bay in close proximity to several CSN sites already included in the evaluation”

p. 3913, line 25: “expected to result in increasingly large fine mode SSA emissions”. Does this refer to quantitatively large emissions or the fine mode emitted SSA is larger in size?

The updated manuscript now includes the following: “For this study, we used Θ values of 30 (consistent with the current CMAQ representation, given as CMAQv5.0.2a or “Baseline”), 20 (CMAQv5.0.2b), 10 (CMAQv5.0.2c), and 8 (CMAQv5.0.2d), which were expected to result in progressively higher emission rates of fine mode SSA (see Figure S1).”

p.3914: Lines15 – 19: In the text and in Table 1 it is unclear how the SST dependence was calculated in CMAQv5.0.2h. Was the third-order dependence of Jaeglé, the linear dependence of Ovadnevaite, or a hybrid used?

The updated manuscript now includes the following: “We conducted two simulations to test the combined effect of setting $\Theta = 8$, SST-dependence, and surf-enhanced emissions (surf zone = 25 meters), with CMAQv5.0.2g using the Jaeglé et al. (2011) third-order SST dependence and CMAQv5.0.2h using a hybrid of the Jaeglé et al. (2011) third-order SST dependence and the Ovadnevaite et al. (2014) process-based linear SST dependence (see Fig. 12 from Ovadnevaite et al. (2014)) for open ocean emissions as follows:”

Table 2: What is the “Corr” term shown? Is it the coefficient of determination, i.e., r^2 ? Also, what are the size ranges of the predicted Aitken and accumulation modes?

Header: Comparison of the mean and Pearson’s correlation coefficient (r) of total observed and model-predicted inorganic particle concentrations ($\mu\text{g m}^{-3}$) at three Bay Regional Atmospheric Chemistry Experiment (BRACE) sites near Tampa, FL.

Footnote: Na^+ predicted for the sum of Aitken and accumulation modes (approximating $\text{PM}_{2.5}$ (Nolte et al., 2015)) and observed for aerosols $< 1.8 \mu\text{m}$ in diameter

p. 3915, lines 13-14: An Aitken and accumulation mode of $D_{p,dry}$ ranging (together) from 10 nm to 1 μm would not result in a direct comparisons with observed concentrations for aerosol with $D_p < 1.8 \mu\text{m}$. In other words, the observations include a significant fraction of the coarse mode not included in the modeled values. At what RH are the diameters that are referred to here?

The updated manuscript now includes the following: “The average fine mode sodium concentration (given as $\text{PM}_{1.8}$ for the measurements and the sum of the Aitken and accumulation approximating $\text{PM}_{2.5}$ (Nolte et al., 2015) for the model predictions) were consistently underpredicted by the Baseline simulation for the BRACE sites with an NMB of -21.6%.”

p. 3916, line 15: What is the peak diameter for a value of 8? This should be stated in the text. For additional clarity, Figure S1 should be moved to the main paper.

The updated manuscript now includes the following: “For the simulations using Θ values ≤ 20 , the lower limit of the SSA dry diameter is decreased to 10 nm to better reflect changes

in the emitted number size distribution (which peaks at ~170, 140, 80, and 60 nm dry diameter for Θ values of 30, 20, 10, and 8 respectively).” Furthermore, we appreciate that the reviewer’s comment on Figure S1 but think that an illustration of an intermediate model development step is more appropriate for the supplement rather than the main text.

Figure 2: It would help guide the eye and compare model and observation results if the observed data were presented as line and markers.

We agree with the reviewer that the observation results could be adjusted to enable comparison with the model predictions. However, the observations did not take place every day of the period and we do not think that connecting these points with a line would be appropriate. In the updated manuscript, we’ve increased the size of the observation points and ordered them above the modeled lines to enable comparison between the two.

Figures 2 and 3: Label the modeled lines as “Revised” and “Baseline” in the figure legend.

These changes have been included in the updated manuscript.

Figure 3: Both model versions overpredict the observed fine + accumulation mode mass concentration of Na. Why? This is not commented on in the text.

The updated manuscript now includes the following: “Both the Baseline and Revised simulations predict a second submicron mode for the three sites that is not evident in the observations; it’s unclear whether this discrepancy is related to inaccuracies in the size-resolved emissions or the modal distribution of the model.”

p. 3917, lines 18 – 20: It is stated that “the Revised simulation well predicted the coarse mode sodium at both the coastal and inland sites.” Based on Figure 3, the Revised simulation over predicts coarse mode Na at the Gandy Bridge site.

The updated manuscript now includes the following: “At the bayside Gandy Bridge site, the very high SST in Tampa Bay results in the well predicted coarse mode sodium in the Baseline simulation becoming overpredicted in the Revised simulation.”

p. 3917, Lines 21 – 23: “Fine mode sodium concentrations increased throughout the BRACE domain in the Revised simulation. . .”. It should be clarified here that the change that is referred to is the difference between the v5.0.2h and v5.0.2a models (at least that is how I interpreted it).

The updated manuscript now includes the following: “Fine (Aitken + accumulation) mode sodium concentrations increased throughout the BRACE domain in the Revised simulation relative to the Baseline simulation...”

p. 3918, lines 22 – 23: Change to “predicted PM2.5 sodium surface concentrations were SLIGHTLY improved in the Revised simulation. . .”

This language has been added to the updated manuscript.

p. 3919, lines 9 – 11: Impacts on sodium from what? Sentence needs to be fixed for clarity.

This sentence has been removed in the updated manuscript.

Figures 5 and 6 (and text): Were modeled PM_{2.5} concentrations used for the comparison with the measurements? Or was the sum of the fine and accumulation modes used? Use of the latter would result in a large underestimation of both sodium and nitrate concentrations.

The updated manuscript now includes the following: “For the CalNex comparison, the sum of the Aitken and accumulation modes was used as the model comparison. However, a comprehensive evaluation of size-resolved inorganic particle composition from Nolte et al. (2015) shows that the difference in the sum of the Aitken and accumulation modes and PM_{2.5} values is generally < 10%.”

p. 3920, lines 2 – 5: It is not surprising that the nitrate underpredictions were not resolved by improved sodium predictions since the sodium concentrations were severely underpredicted even in the Revised simulations.

The updated manuscript now includes the following: “In Riverside, for example, nitrate underpredictions in the Revised simulation were likely due to a combination of persistent sodium underpredictions and an underestimate of ammonia emissions from upwind dairy facilities (Nowak et al., 2012; Kelly et al., 2014).”