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Updating sea spray aerosol emissions in the Community Multiscale Air Quality (CMAQ) model version 5.0.2

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Abstract

Sea spray aerosols (SSA) impact the particle mass concentration and gas-particle partitioning in coastal environments, with implications for human and ecosystem health. Despite their importance, the emission magnitude of SSA remains highly uncertain with global estimates varying by nearly two orders of magnitude. In this study, the Community Multiscale Air Quality (CMAQ) model was updated to enhance fine mode SSA emissions, include sea surface temperature (SST) dependency, and reduce coastally-enhanced emissions. Predictions from the updated CMAQ model and those of the previous release version, CMAQv5.0.2, were evaluated using several regional and national observational datasets in the continental US. The updated emissions generally reduced model underestimates of sodium, chloride, and nitrate surface concentrations for an inland site of the Bay Regional Atmospheric Chemistry Experiment (BRACE) near Tampa, Florida. Including SST-dependency to the SSA emission parameterization led to increased sodium concentrations in the southeast US and decreased concentrations along parts of the Pacific coast and northeastern US. The influence of sodium on the gas-particle partitioning of nitrate resulted in higher nitrate particle concentrations in many coastal urban areas due to increased condensation of nitric acid in the updated simulations, potentially affecting the predicted nitrogen deposition in sensitive ecosystems. Application of the updated SSA emissions to the California Research at the Nexus of Air Quality and Climate Change (CalNex) study period resulted in modest improvement in the predicted surface concentration of sodium and nitrate at several central and southern California coastal sites. This SSA emission update enabled a more realistic simulation of the atmospheric chemistry in environments where marine air mixes with urban pollution.

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1 Introduction

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 (Pierce and Adams, 2006). The chemical composition of SSA (e.g., major ions: Na^+ , Mg^{2+} , Ca^{2+} , K^+ , Cl^- , SO_4^{2-} ; Tang et al., 1997) is affected by atmospheric processing, with the uptake of nitric acid (Gard et al., 1998, and references therein), sulfuric acid (McInnes et al., 1994), dicarboxylic acids (Sullivan and Prather, 2007), and methylsulfonic acid (Hopkins et al., 2008) shown to be important processes. Sea spray aerosols also influence gas-phase atmospheric chemistry via displacement of chlorine and bromine from the particle phase and subsequent impacts on ozone formation and destruction (Yang et al., 2005; Long et al., 2014). Despite this importance, emissions of sea spray aerosols are poorly constrained with global estimates ranging from 2 to 120 Pgyr⁻¹ (de Leeuw et al., 2011).

An active area of recent research has been in the determination of the SSA size distribution. The size distribution of particles influences their atmospheric lifetime, surface area available for heterogeneous reactions, cloud condensation nuclei efficiency, and optical properties. A widely-used SSA emission parameterization in early chemical transport models was described by Monahan et al. (1986) which predicts the size distribution between 0.8 and 8 μm in dry diameter based on laboratory measurements. To address the overpredicted SSA emission rate when Monahan et al. (1986) parameterization was extended to aerosol dry diameters $< 0.2 \mu\text{m}$ (Andreas, 1998; Vignati et al., 2001), Gong (2003) revises the Monahan et al. (1986) parameterization to match the SSA size distribution observed in the North Atlantic (O'Dowd et al., 1997) down to 0.07 μm dry diameter. Since the publication of Gong (2003), several studies have examined the size distribution of SSA generated in the laboratory and measured in field campaigns (Mårtensson et al., 2003; Clarke et al., 2006; Sellegri et al., 2006; Keene et al., 2007; Tyree et al., 2007; Norris et al., 2008; Fuentes et al., 2010). In a review of SSA emission measurements from both laboratory- and field-based studies, de Leeuw

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et al. (2011) shows a broad range (0.05–0.1 μm in dry diameter) of particle sizes having the maximum number production flux. Recent SSA production parameterizations (see Grythe et al., 2014) reflect these measurements, with most having a production rate maximum at aerosol sizes lower than the lower cutoff (0.07 μm dry diameter) of Gong (2003). Due to the lack of detailed submicron measurements at the time, the Gong (2003) parameterization was given as:

$$\frac{dF}{dr} = 1.373U_{10}^{3.41}r^{-(4.7(1+\Theta)r^{-0.017}r^{-1.44})}(1 + 0.057r^{3.45}) \times 10^{1.607}e^{-((0.433-\log r)/0.433)^2} \quad (1)$$

where $\frac{dF}{dr}$ is the SSA number flux with units of $\text{m}^{-2}\text{s}^{-1}\mu\text{m}^{-1}$, r is the particle radius in μm at 80 % relative humidity, U_{10} is the 10 m wind speed in m s^{-1} , and Θ is an adjustable shape parameter that controlled the submicron size distribution. Gong (2003) tested Θ values between 15 and 40, suggesting (with limited observational evidence) a Θ value of 30.

Seawater temperature can increase or decrease SSA number emissions by up to ~ 100 % due to the temperature dependency of surface tension, density, viscosity, and air entrainment (Mårtensson et al., 2003; Sellegri et al., 2006; Zábori et al., 2012a; Ovadnevaite et al., 2014; Callaghan et al., 2014). Mårtensson et al. (2003), Sellegri et al. (2006), and Zábori et al. (2012a) all observe a negative temperature dependence for the production flux of SSA < 70 nm diameter in synthetic seawater laboratory experiments. Similar negative temperature dependencies are measured in SSA generated from Arctic Ocean seawater (Zábori et al., 2012b). Mårtensson et al. (2003) and Sellegri et al. (2006) also reported positive temperature dependencies for the SSA production flux for particles larger than 70 nm in diameter. This difference in the temperature-dependence of small and large SSA emissions is likely due to their bubble size-dependence and impact of SST on small and large bubbles (Sellegri et al., 2006). Sofiev et al. (2011) develops a size-dependent temperature correction factor for SSA emissions reflecting the different temperature dependencies of fine and coarse mode aerosols. A global comparison of observed and model predicted coarse mode

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sea salt concentrations in Jaeglé et al. (2011) leads to the development of a third order polynomial function for the SST dependence of the Gong et al. (2003) SSA emission parameterization. Grythe et al. (2014) compares the Jaeglé et al. (2011) and Sofiev et al. (2011) temperature dependencies, finding that the Jaeglé et al. (2011) function gives the best model improvement to the observed temperature dependence. Modeling studies implementing the Jaeglé et al. (2011) temperature-dependent SSA emissions have shown improved prediction of surface sea-salt mass concentration (Spada et al., 2013; Grythe et al., 2014) relative to temperature-independent emissions. Using a process-based approach incorporating seawater viscosity and wave state, Ovadnevaite et al. (2014) finds a positive temperature dependence of SSA emissions similar to Jaeglé et al. (2011) but resembling a linear (rather than third order polynomial) relationship.

In addition to bubble bursting in the open ocean, SSA can be emitted via wave breaking in the surf zone covering an area roughly 20 to 100 m from the coastline (Petelski and Chomka, 1996; Lewis and Schwartz, 2004). Surf zone SSA emissions have been shown to be enhanced relative to the open ocean, resulting in higher sea-salt concentrations near the coast (de Leeuw et al., 2000). Vignati et al. (2001) concludes that surf zone SSA emissions provide additional surface for heterogeneous reactions and impact the atmospheric chemistry of coastal areas. There are limited observations and large uncertainties in the surf zone SSA emissions related to the zone width and whitecap coverage, with de Leeuw et al. (2000) observing a 30 m wide surf zone with an assumed 100 % whitecap fraction on the California coast and Clarke et al. (2006) observing a mean whitecap fraction in the 35 m wide surf zone of 40 % in Hawaii. The inclusion of surf zone emissions increases sodium and chloride concentrations by a factor of 10 and improves the predicted concentration of particulate matter (PM) < 10 μm in diameter (PM₁₀) by up to 20 % in the Eastern Mediterranean (Im, 2013).

The current SSA treatment in the Community Multiscale Air Quality (CMAQ) model version 5.0.2 is described by Kelly et al. (2010) and includes the open ocean emissions of Gong (2003), coastally-enhanced emissions similar to de Leeuw et al. (2000)

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in which a fixed whitecap coverage of 100 % is applied to the Gong (2003) parameterization for a 50 m-wide surf zone, and dynamic transfer of HNO_3 , H_2SO_4 , HCl , and NH_3 between coarse mode particles and the gas phase. Based on comparison with observations from three Tampa, Florida sites at different distances from the coastline, Kelly et al. (2010) finds that enhancing sea spray emissions in coastal grid cells according to a 50 m wide surf zone with a 100 % whitecap coverage improved CMAQ model underprediction of sodium, chloride, and nitrate concentrations (particularly at the coastal site) relative to a simulation with only the Gong (2003) open ocean emissions. The dynamic transfer of HNO_3 , H_2SO_4 , HCl , and NH_3 between coarse particles and the gas phase as implemented by Kelly et al. (2010) further improves predicted concentrations of semi-volatile species like chloride and nitrate. Despite these improvements, persistent underpredictions of sodium, chloride, and nitrate concentrations at the inland site remain unresolved. In this work, we expand upon the Kelly et al. (2010) CMAQ SSA emission treatment by updating the fine mode size distribution, SST dependence, and coastally-enhanced emissions to reflect recent SSA research. Due to the advanced treatment of SSA chemistry in CMAQ, their emissions can be evaluated using concentrations of the directly-emitted sea-salt components such as sodium and species such as nitrate that react with sea-salt components in the atmosphere. Specifically, we hypothesize that the improved prediction of sodium will correspond to improvements in the gas-particle partitioning of nitrate aerosol as suggested by Kelly et al. (2014). The goal of this work is to improve the size distribution, magnitude, and spatiotemporal variability of CMAQ-predicted SSA emissions and the resulting impacts on atmospheric chemistry in coastal and inland areas.

2 Methods

2.1 Observational datasets

Two field campaigns with different meteorology, atmospheric chemistry, and SSA sources from oceans having distinct surface temperatures and bathymetry were used to evaluate the updated emissions. The Bay Regional Atmospheric Chemistry Experiment (BRACE) (Atkeson et al., 2007; Nolte et al., 2008) was conducted from May to June 2002 at three sites (Azalea Park: 27.78° N, 82.74° W, Gandy Bridge: 27.89° N, 82.54° W, and Sydney: 27.97° N, 82.23° W) around Tampa Bay, FL (see Fig. 1). These three sites represent coastal (Azalea Park), bayside (Gandy Bridge) and inland (Sydney) regions, and roughly 1, 25, and 50 km from the Gulf of Mexico coastline. Size-resolved measurements of inorganic PM composition were made with four micro-orifice cascade impactors, which operated for 23 h per sample (Evans et al., 2004). The cascade impactors had 8–10 fractionated stages ranging from 0.056 to 18 μm in aerodynamic diameter, and two cascade impactors were collocated at the Sydney site. Additionally, particulate nitrate and nitric acid were measured at a high temporal resolution (≤ 15 min) using a soluble particle collector employing ion chromatography (Dasgupta et al., 2007) and denuder difference (Arnold et al., 2007).

The California Research at the Nexus of Air Quality and Climate Change (CalNex) 2010 field project was conducted from May to July 2010 throughout California. The goal of the study was to simultaneously measure variables affected by emissions, atmospheric transport and dispersion, atmospheric chemical processing, and cloud-aerosol interactions and aerosol radiative effects (Ryerson et al., 2013). The South Coast portion of the CalNex campaign included continuous ground-based measurements of PM < 2.5 μm in diameter (PM_{2.5}) composition using particle-into-liquid sampling and ion chromatography (Weber et al., 2001) and the mixing ratio of many gases at Pasadena, CA (34.14° N, 118.12° W, ~ 35 km from the Pacific coast). Here, we evaluated CMAQ using surface concentrations of sodium and nitrate measured continuously at Pasadena and as daily averages every three days at sites operated by the Chem-

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ical Speciation Network (CSN) within the South Coast, San Francisco Bay, and San Diego air basins. Hereafter, these CSN sites and the Pasadena site will collectively be referred to as the coastal CalNex sites.

In addition to local field campaigns, we evaluated SSA emissions in CMAQ against surface $PM_{2.5}$ concentrations of sodium and nitrate measured throughout the continental US (CONUS) as part of the Interagency Monitoring of Protected Visual Environments (IMPROVE) for remote/rural locations and CSN for urban locations during the May 2002 BRACE time period. Daily-average sodium mass concentrations in the IMPROVE and CSN networks were measured once every three days via tube-generated X-ray fluorescence (XRF) (White, 2008). Nitrate concentrations for both the IMPROVE and CSN networks are determined by ion chromatography. During the May 2002 period, the IMPROVE network consisted of ~ 160 sites while the CSN network consisted of ~ 230 sites.

2.2 Model configuration

In this work, we used the CMAQ model v5.0.2 to simulate the impact of updated sea spray aerosol emissions on surface aerosol concentrations/size distribution and gas-particle partitioning. CMAQ represents the aerosol size distribution using three modes (Aitken, accumulation, and coarse) and simulates inorganic aerosol thermodynamics using ISORROPIA II (Binkowski and Roselle, 2003; Fountoukis and Nenes, 2007). Kelly et al. (2010) further enhanced the SSA chemical treatment in CMAQ by allowing dynamic transfer of HNO_3 , H_2SO_4 , HCl, and NH_3 between coarse particles and the gas phase. For comparison with the CONUS observational datasets such as IMPROVE and CSN, we used a model domain covering the continental US at 12 km \times 12 km horizontal resolution and 41 vertical layers with a surface layer up to 20 m a.g.l. The simulation time period (1 May to 3 June 2002 with an 11 day spin-up) was made to coincide with the BRACE campaign to enable additional evaluation of the coastal-to-inland changes in the aerosol composition/size distribution and gas-particle partitioning. Meteorological parameters were generated by the Weather Research Forecasting model (WRF) ver-

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emission parameterizations (Grythe et al., 2014). The radius of peak emissions at 80 % relative humidity (RH) from the Gong (2003) parameterization with a Θ value of 8 was ~ 60 nm; this value was similar to the radius of maximum production flux from several parameterizations reviewed in de Leeuw et al. (2011).

Including the positive temperature dependence for SSA emissions in CMAQ was expected to affect the seasonality and spatial distribution of predicted concentrations. The Jaeglé et al. (2011) third order polynomial function of SST dependence for SSA emissions (CMAQv5.0.2e) increases the summertime/tropical concentrations, decreases wintertime/polar concentrations, and leaves mid-latitude/spring/autumn concentrations relatively unchanged. The surf zone width used in parameterizing the coastal enhancement of emissions was decreased from 50 to 25 m (CMAQv5.0.2f), reflecting both the uncertainty in the width distance and whitecap fraction within the surf zone. As SSA emissions from coastal grids impact a narrow region, adjusting the surf zone width was expected to strongly affect coastal concentrations while having a relatively minor effect on downwind concentrations. We conducted two simulations to test the combined effect of setting $\Theta = 8$, SST-dependence, and coastally-enhanced emissions (surf zone = 25 m), with CMAQv5.0.2g using the Jaeglé et al. (2011) third-order SST dependence and CMAQv5.0.2h using the adapted Ovadnevaite et al. (2014) process-based linear SST dependence for open ocean emissions as follows:

$$\frac{dF}{dr} = (0.38 + 0.054 \times \text{SST}) \times 1.373 U_{10}^{3.41} r^{-(4.7(1+8r)^{-0.017r^{-1.44}})} (1 + 0.057 r^{3.45}) \times 10^{1.607 e^{-(0.433 - \log r)/0.433}^2} \quad (2)$$

where SST has units of $^{\circ}\text{C}$. The updated SSA emission parameterization given in Eq. (2) was mapped to the CMAQ aerosol modes as a function of relative humidity following Zhang et al. (2005, 2006). A summary of the different CMAQ model simulations in which SSA emissions were changed is given in Table 1. The approach used in CMAQv5.0.2h is planned to be included in the next public release of CMAQ (version 5.1).

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3 Results

3.1 BRACE

The total particulate (PM_{tot}) nitrate, chloride, and sodium concentrations observed at the three sites during the BRACE campaign and corresponding CMAQ predicted concentrations for the “Baseline” (v5.0.2a) and sensitivity simulations (v5.0.2b–h) are summarized in Table 2. Generally, the Baseline simulation underpredicted the nitrate concentrations for all sites with a normalized mean bias (NMB) of -46.4% . The Baseline simulation predicted the magnitude of chloride and sodium at the coastal site (Azalea Park) relatively well. However, it increasingly underpredicted chloride and sodium as the distance from the shore increased. The Baseline simulation overestimated by approximately a factor of 2 the observed decrease in PM_{tot} chloride and sodium between the coastal Azalea Park and inland Sydney sites. The average fine mode sodium concentration (given as $PM_{1.8}$ for the measurements and the sum of the Aitken and accumulation modes for the model predictions) were consistently underpredicted by the Baseline simulation for the BRACE sites with an NMB of -21.6% . As the Θ value was changed from 30 to 20 (v5.0.2b), the predicted PM_{tot} chloride and sodium (and nitrate via secondary processes) decreased slightly despite an increase in fine mode sodium concentrations. This surprising result was due to slight differences in the fitting of coarse mode SSA emissions to CMAQ’s aerosol modes. The transition of Θ values from 20 to 10 to 8 led to small ($< 10\%$) increases in the nitrate, chloride, and sodium concentrations relative to the Baseline simulation for all sites. Although it slightly overestimated chloride and sodium at the coastal Azalea Park site, the v5.0.2d simulation with a Θ value of 8 had the best prediction (both in terms of magnitude and correlation) of concentrations at the Gandy Bridge and Sydney sites.

The modeled chloride and sodium aerosol concentrations were much more sensitive to the implementation of SST-dependent SSA emissions (v5.0.2e) and reduction of the surf zone width used for coastal SSA emission enhancement (v5.0.2f) than the changing of the Θ values. With the positive temperature dependence of the Jaeglé

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veloped for GEOS-Chem in Jaeglé et al. (2011). Therefore, the v5.0.2h simulation is referred hereafter as the “Revised” simulation.

The statistical improvement in the Revised simulation relative to the Baseline (v5.0.2a) simulation is reflected in the time series of sodium concentrations at the three sites (Fig. 2). Besides showing the generally higher PM_{tot} sodium concentrations at the bayside and inland sites and higher $PM_{1.8}$ sodium concentrations at all sites, Fig. 2 also shows that the Revised simulation diverges most from v5.0.2a during periods of high SSA concentration episodes (15, 22 May 2002). This suggests that the Revised simulation better replicated the sea spray aerosol emissions during periods with strong onshore flow compared to the Baseline simulation. The range of $PM_{1.8}$ sodium concentrations predicted by the Revised simulation was more consistent with observations than the Baseline simulation, especially at the Sydney site which has observed concentrations of $0.05\text{--}0.27\ \mu\text{g m}^{-3}$ and predicted concentrations of $0.02\text{--}0.16\ \mu\text{g m}^{-3}$ and $0.03\text{--}0.25\ \mu\text{g m}^{-3}$ for the Baseline and Revised simulations. Comparison of the predicted and observed size distribution of sodium at the three sites (see Fig. 3) showed that much of the observed and predicted decrease in the sodium mass concentration in the transition from coastal to inland sites occurred within the coarse mode. The Baseline simulation overpredicted/underpredicted coarse mode sodium at the coastal/inland sites, while the Revised simulation well predicted the coarse mode sodium at both the coastal and inland sites.

Fine (Aitken + accumulation) mode sodium concentrations increased throughout the BRACE domain in the Revised simulation with larger changes (up to $0.1\ \mu\text{g m}^{-3}$) offshore and smaller changes ($0.05\ \mu\text{g m}^{-3}$) inland as shown in the right column of Fig. 1a. The total (sum of Aitken, accumulation, and coarse modes) sodium concentrations over the open ocean increased in the warmer southern waters of the Atlantic and Pacific Oceans and decreased in the cooler waters off New England and the Pacific Northwest. Grid cells directly adjacent to the coast experienced concentration decreases of up to $1\ \mu\text{g m}^{-3}$, with the largest decreases occurring for cells with large surf zones due to irregular coastlines (i.e. barrier islands, peninsulas, etc). These coastline-centered

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decreases were limited spatially, as adjacent cells just offshore had large increases in sodium concentration. Like the fine mode changes, the largest total sodium concentration increases occurred offshore while more modest increases were predicted for inland locations. The coastal-inland concentration gradients were stronger for the total concentration changes due to the faster deposition velocity of coarse mode aerosols (relative to the fine mode) which comprise most of the total mass.

The hourly time series of observed and predicted nitrate gas/particle partitioning from the Sydney site for May 2002 (Fig. 4) shows that the Revised simulation pushes the partitioning towards the particle phase (relative to the Baseline simulation) and closer to observations. The average observed fraction of nitrate in the particle phase was 0.51 while the predicted fractions from the Baseline and Revised simulations were 0.36 and 0.42, respectively. Figure 4 indicates that the largest difference in the nitrate partitioning between the Baseline and Revised simulations occurred during the daytime, when higher concentrations of inorganic ions like sodium prevented some of the nitric acid evaporation from the particle phase during the hot afternoon period. Despite improvement in the daytime partitioning, the Revised simulation continued to overpredict the nighttime nitrate fraction and daytime nitric acid fraction. This impact on partitioning is consistent with Kelly et al. (2014), which suggested that improving CMAQ prediction of sodium concentration and relative humidity would improve gas-particle partitioning of nitrate in the CalNex model domain.

3.2 CalNex

Similar to results for the BRACE sites, the predicted $PM_{2.5}$ sodium surface concentrations were improved in the Revised simulation relative to the Baseline for sites examined during the CalNex simulation period (see Fig. 5). Surface sodium concentrations were underpredicted by both the Baseline and Revised simulations for all the coastal CalNex sites, especially in the 11–16 June time period when high sodium concentrations at several of the sites were not well captured by either the Revised or Baseline simulation. It is worth noting that a sensitivity test in which the coastally-enhanced

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NaNO₃ stoichiometry would lead to a greater increase of nitrate than sodium mass. The nitrate underpredictions in Fig. 6 were not resolved entirely by improved sodium predictions. In Riverside, for example, nitrate underpredictions in the Revised simulation were likely due in part to underestimates of ammonia emissions from upwind dairy facilities (Nowak et al., 2012; Kelly et al., 2014).

3.3 Continental US

Unlike the PM_{1.8} or PM_{2.5} sodium concentrations evaluated using the BRACE and CalNex observations, the total sodium surface concentration changes shown in Fig. 1b both increased and decreased in the CONUS domain due to the variability in coastal and oceanic SST. The distribution of fine (Aitken + accumulation) mode concentration changes (Fig. 1a) had some similar features to the total concentration changes (Fig. 1b), with the largest increases occurring over areas with high (>~ 20 °C) SSTs. Differences between the fine mode and total concentration changes were most notable for regions with low (<~ 10 °C) SSTs (Pacific and northeast US coasts) and for inland regions. Because fine mode particles have a low dry deposition velocity, offshore increases in the fine mode sodium concentrations were able to extend inland and lead to increased deposition (see Fig. S4a). The flat topography and large offshore concentration increases in the southeast US resulted in concentration increases of up to 0.25 μg m⁻³ hundreds of kilometers from the coast. While reductions in fine mode SSA emissions due to low SSTs were balanced by increased emissions from changing Θ, cold seawater temperatures off the Pacific coast and northeast US led to large decreases in total sodium concentration of up to -0.5 μg m⁻³. As in the BRACE domain, the decrease in coastally-enhanced emissions led to localized decreases in PM_{tot} sodium concentration for grid cells immediately adjacent to the coastline throughout the CONUS domain. Regions with rugged coastlines and barrier islands experienced the largest concentration decreases because of the large surf zone area.

Model comparison of PM_{2.5} sodium concentrations from the IMPROVE and CSN networks revealed improvement from the Baseline to Revised simulation (see Fig. 7). For

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creased deposition of total nitrate inland because of the lower dry deposition velocity of nitrate aerosol relative to nitric acid (see Fig. S4b).

4 Conclusions

In this study, the size distribution, temperature dependence, and coastal enhancement of sea spray aerosol (SSA) emissions were updated in the Community Multiscale Air Quality (CMAQ) model version 5.0.2. Increasing fine mode emissions, including temperature dependence, and reducing the coastally-enhanced emissions from the “Baseline” to the “Revised” simulation collectively improved the summertime surface concentration predictions for sodium, chloride, and nitrate at three Bay Regional Atmospheric Chemistry Experiment (BRACE) sites near Tampa, Florida. Surface concentrations at the inland site near Tampa were particularly affected by these emission changes, as low dry deposition velocities for the fine mode aerosols increased the atmospheric lifetime and inland concentrations. The coastal-inland concentration gradient was also affected by the updated emissions, as the reduction in surf zone width used to enhance coastal emissions brought the Revised simulation in closer agreement with observations. These SSA emission updates led to increases in the fine mode sodium surface concentrations throughout coastal areas of the continental US, with the largest increases occurring near the Southeast US coast where sea surface temperatures (SST) were high. Decreases in the total sodium concentration were predicted for oceanic regions with low SST such as the Pacific and northern Atlantic coasts. Comparison of the Baseline and Revised simulation with sodium observations from the IMPROVE and CSN networks showed that the updated emissions reduced the widespread underprediction of concentrations, especially in the Southeast and mid-Atlantic US. Non-linear responses between changes in total and sea-salt $PM_{2.5}$ concentrations indicated that the impacts of these emissions changes on aerosol chemistry were enhanced in polluted coastal environments. The Revised simulation had increased sodium and nitrate

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aerosol concentrations at most CalNex sites, slightly reducing the underprediction from the Baseline simulation.

Potential future work includes treating the organic fraction of SSA (Gantt et al., 2010), implementing the Group for High Resolution Sea Surface Temperature (GHR SST) dataset (Donlon et al., 2007), and linking the SSA emissions to marine boundary layer halogen chemistry via debromination (Yang et al., 2005). Episodic high SSA concentrations are not well captured at any of the coastal CalNex sites in the Revised simulation, suggesting that other factors not accounted for in our updated SSA emission parameterization such as wind history, wave state, ocean biology, solar radiation, whitecap timescales, or the limited ocean surface area in the modeling domain (Callaghan et al., 2008, 2014; Ovadnevaite et al., 2014; Long et al., 2014) may play an important role. Additional model developments focused on the South Coast region of California are warranted considering the impact on nitrate discussed above as well as the impact that reactive chlorine atoms derived from sea spray particles can have on ozone in this region (Simon et al., 2009; Sarwar et al., 2012; Riedel et al., 2014). As the fine mode size distribution has a far greater impact on the number concentration than the mass concentration, the changes described in this study likely impact other model parameters such as aerosol radiative feedbacks which are included in the coupled WRF-CMAQ modeling system (Gan et al., 2014).

Code availability

The updated code is available upon request prior to the public release of CMAQ v5.1. Please contact Jesse Bash at Bash.Jesse@epa.gov for more information.

The Supplement related to this article is available online at doi:10.5194/gmdd-8-3905-2015-supplement.

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Table 1. Differences in CMAQ model version used in this study.

Model	Θ	SST-dependence	Surf Zone (m)
CMAQv5.0.2a ^a	30	NA	50
CMAQv5.0.2b	20	NA	50
CMAQv5.0.2c	10	NA	50
CMAQv5.0.2d	8	NA	50
CMAQv5.0.2e	30	Jaeglé et al. (2011)	50
CMAQv5.0.2f	30	NA	25
CMAQv5.0.2g	8	Jaeglé et al. (2011)	25
CMAQv5.0.2h ^b	8	Jaeglé et al. (2011); Ovadnevaite et al. (2014)	25

^a This simulation is also referred to as the “Baseline” simulation.

^b In this simulation, which is also referred to as the “Revised” simulation, the SST-dependence of Jaeglé et al. (2011) has been linearized following Ovadnevaite et al. (2014).

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Table 2. 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.

Species	Obs.	v5.0.2a		v5.0.2b		v5.0.2c		v5.0.2d		v5.0.2e		v5.0.2f		v5.0.2g		v5.0.2h	
		Mean	Corr														
Azalea Park																	
NO_3^-	1.96	0.74	0.34	0.72	0.33	0.73	0.34	0.76	0.35	0.92	0.30	0.65	0.45	0.74	0.45	0.79	0.43
Cl^-	1.93	2.41	0.17	2.33	0.15	2.36	0.15	2.49	0.18	3.69	0.19	1.55	0.31	1.92	0.38	2.15	0.42
Na^+	1.62	1.62	0.19	1.61	0.18	1.62	0.18	1.71	0.21	2.39	0.22	1.11	0.33	1.38	0.41	1.52	0.44
Na^{**}	0.13	0.11	0.38	0.16	0.42	0.15	0.41	0.16	0.42	0.15	0.42	0.10	0.43	0.16	0.53	0.18	0.58
Gandy Bridge																	
NO_3^-	1.74	1.32	0.55	1.03	0.54	1.03	0.54	1.07	0.55	1.32	0.51	0.93	0.60	1.09	0.61	1.17	0.61
Cl^-	1.72	1.57	0.71	1.51	0.71	1.53	0.71	1.63	0.71	2.53	0.68	1.32	0.81	1.91	0.81	2.26	0.81
Na^+	1.46	1.17	0.67	1.17	0.67	1.17	0.67	1.24	0.67	1.78	0.65	1.01	0.79	1.41	0.81	1.62	0.80
Na^{**}	0.13	0.09	0.51	0.13	0.54	0.12	0.53	0.13	0.54	0.12	0.51	0.09	0.56	0.14	0.60	0.17	0.63
Sydney																	
NO_3^-	1.51	0.73	0.58	0.71	0.57	0.72	0.57	0.75	0.58	0.88	0.59	0.68	0.60	0.78	0.63	0.84	0.64
Cl^-	1.31	0.82	0.35	0.78	0.35	0.79	0.35	0.86	0.36	1.32	0.30	0.71	0.49	1.02	0.50	1.26	0.53
Na^+	1.14	0.67	0.44	0.66	0.45	0.67	0.45	0.72	0.46	0.98	0.41	0.59	0.55	0.82	0.57	0.98	0.61
Na^{**}	0.11	0.09	0.19	0.12	0.27	0.11	0.25	0.12	0.27	0.11	0.21	0.08	0.23	0.13	0.33	0.16	0.40

* Na^+ predicted for the sum of Aitken and accumulation modes and observed for aerosols < 1.8 μm in diameter.

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Table 3. Statistical comparison between observed and model-predicted sodium, nitrate and $\text{PM}_{2.5}$ surface concentrations ($\mu\text{g m}^{-3}$) for the continental US in May 2002 from the IMPROVE and CSN networks.

Species	Obs.	v5.0.2a		v5.0.2g		v5.0.2h	
		Mean	Corr	Mean	Corr	Mean	Corr
IMPROVE							
Na^+	0.44	0.16	0.11	0.16	0.17	0.19	0.20
NO_3^-	0.61	0.23	0.28	0.26	0.26	0.26	0.27
$\text{PM}_{2.5}$	5.98	4.24	-0.04	4.16	-0.01	4.30	0.04
CSN							
Na^+	0.34	0.11	0.59	0.14	0.62	0.15	0.62
NO_3^-	1.94	0.61	0.76	0.68	0.76	0.68	0.75
$\text{PM}_{2.5}$	9.74	6.04	0.74	6.29	0.74	6.48	0.74

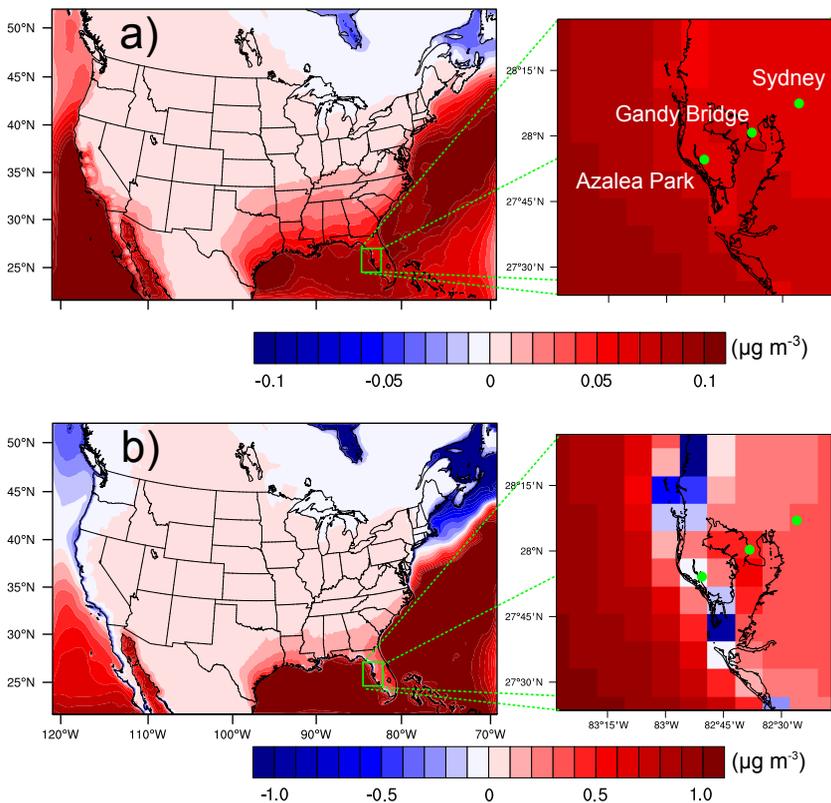


Figure 1. Change in the (a) fine mode and (b) total surface sodium concentration between the CMAQv5.0.2h and CMAQv5.0.2a simulations for May 2002 over the continental US and BRACE domains with sites from left to right of Azalea Park, Gandy Bridge, and Sydney as green dots. Sodium concentrations are consistently greater for the fine mode in the CMAQv5.0.2h simulation and are greater or less depending on location for the total concentration.

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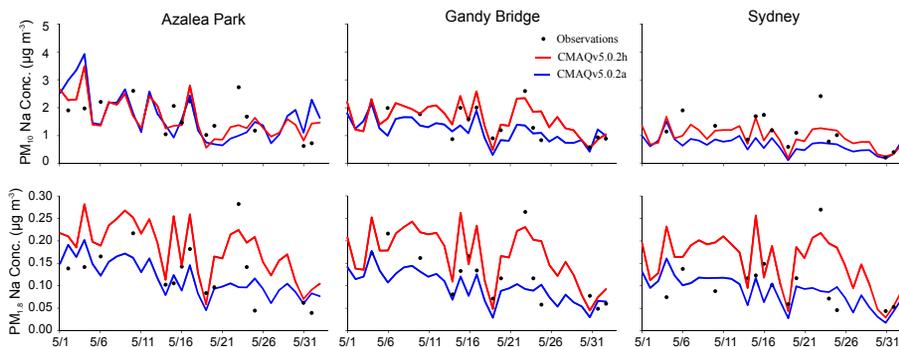


Figure 2. Time series of the observed and predicted daily PM_{10} and $PM_{1.8}$ Na^+ concentration at the three BRACE sites. Note that the $PM_{1.8}$ Na^+ concentration predicted by CMAQ is represented by the sum of the Aitken and accumulation modes.

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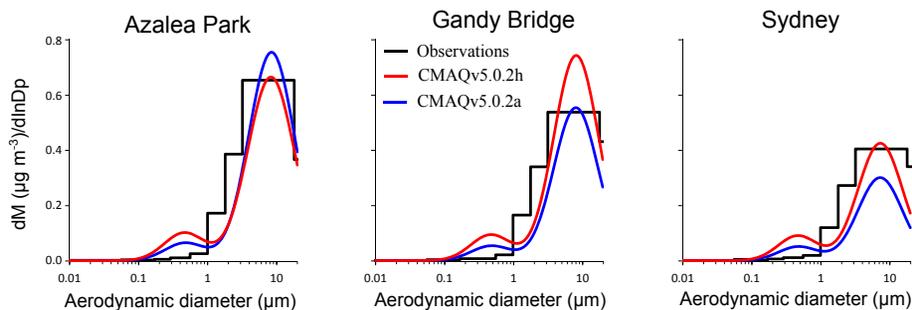


Figure 3. Observed and predicted size distributions of Na^+ at the three Tampa-area sites averaged over 15 sampling days (14 at Sydney) during 2 May–2 June 2002.

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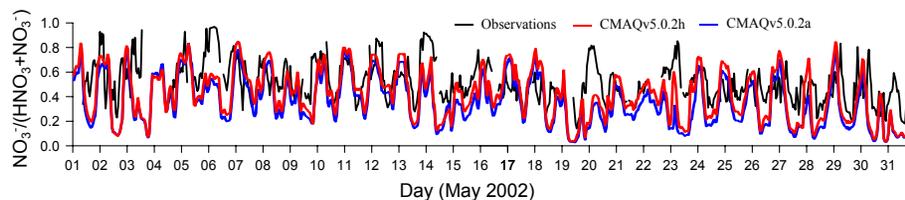


Figure 4. Time series of observed and modeled fraction of total nitrate in the particle phase $[\text{NO}_3^-/(\text{HNO}_3 + \text{NO}_3^-)]$ at the Sydney, FL site for May 2002. Tick marks represent 00:00 local standard time on each day.

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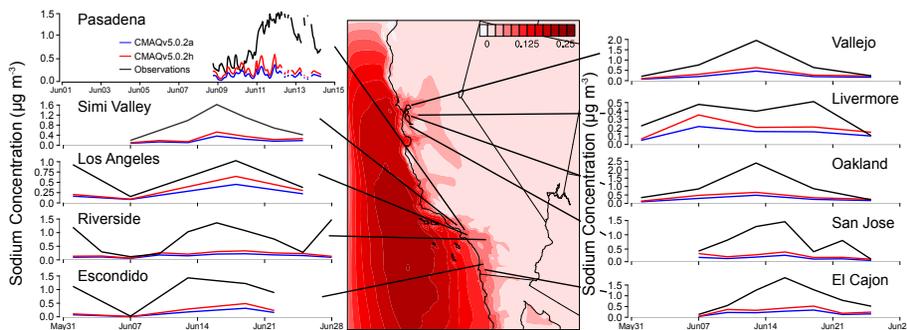


Figure 5. Change ($\mu\text{g m}^{-3}$) in the fine (Aitken + accumulation) mode surface sodium concentration between the CMAQv5.0.2h and CMAQv5.0.2a simulations for June 2010 over the CalNex domain surrounded by time series plots of the observed and predicted daily and/or hourly $\text{PM}_{2.5}$ sodium concentration at the coastal CalNex sites.

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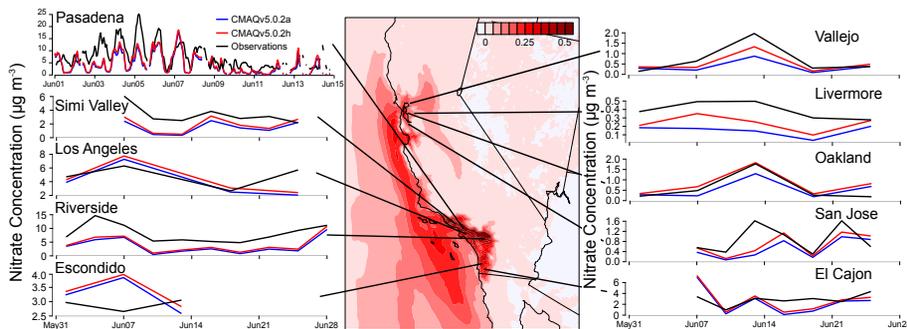


Figure 6. Change ($\mu\text{g m}^{-3}$) in the fine (Aitken + accumulation) mode surface nitrate concentration between the CMAQv5.0.2h and CMAQv5.0.2a simulations for June 2010 over the CalNex domain surrounded by time series plots of the observed and predicted daily and/or hourly $\text{PM}_{2.5}$ nitrate concentration at the coastal CalNex sites.

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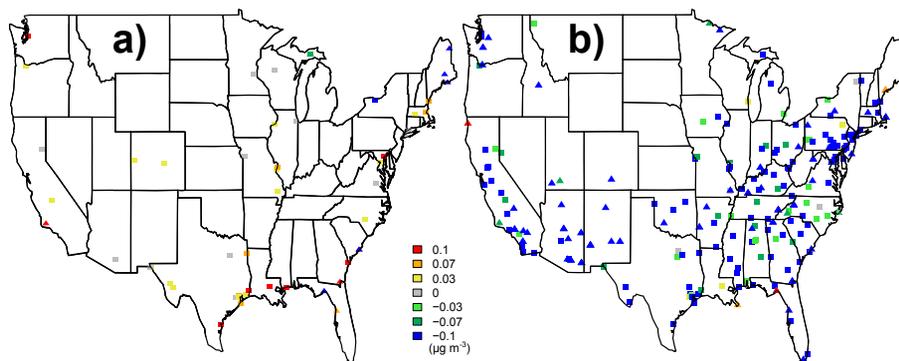


Figure 7. Model bias of $\text{PM}_{2.5}$ -sodium concentration predicted by the CMAQv5.0.2h simulation compared to observations from the IMPROVE (triangles) and CSN (squares) networks for May 2002 segregated by an (a) increase or (b) decrease in the error relative to the CMAQv5.0.2a simulation. The map only includes data where the model percentage difference between the CMAQv5.0.2a and CMAQv5.0.2h simulations is $> 5\%$.

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