Observationally constrained analysis of sea salt aerosol in the marine atmosphere

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Supplementary Material

Three sea salt emission algorithms used in GEOS GOCART

A lot of effort has been devoted to develop and improve the parameterization of sea salt emission algorithms, which are primarily dependent on wind speed, atmospheric stability, sea surface and air temperatures, and salinity in the near-surface ocean waters (Lewis and Schwartz, 2004; Barthel et al., 2014). Here we examine three of the sea salt emission schemes that have been implemented in GEOS/GOCART: 1) a scheme proposed by Gong (2003) (named Emi1), 2) a derivative of Gong's parameterization with a modified wind source term developed at the NASA Global Modeling and Assimilation Office (GMAO) using Moderate Resolution Imaging Spectroradiometer (MODIS) AOD over vast oceans (Emi2), and 3) the sea salt emission scheme used in the Modern-Era Retrospective Analysis for Research and Applications version 2 (MERRA2) reanalysis that has the same wind source function as Emi2 but also includes a sea surface temperature correction tailored to GEOS (Emi3). Emi1 is the sea salt emission that is widely used by aerosol community (Jaeglé et al. 2011; Spada et al., 2015; Textor et al., 2006). Emi3, which is derived based on Emi1 and Emi2, is the current default sea salt emission used by GEOS GOCART in the sea salt simulations presented in the main body of this paper. ATom global surveys, by providing comprehensive and independent sea salt observations, allow us to confirm that the Emi3 is indeed an improved emission.

Details for Emi2 and Emi3 are described here. Sea salt emissions are controlled by aerosol particles generated from collapsing bubbles and ejected jet droplets that in turn are directly related to the whitecap fraction in the ocean and are commonly parameterized as a function of wind speed and SST. In global models this functional dependence is further simplified and expressed as the product of 10-m wind ($W(u_{10m})$), SST ($T(SST)$), and size distribution terms ($S(D)$), e.g., number flux:

$$\frac{dN}{dD} = W(u_{10m})\, T(SST)\, S(D)$$

The $W(u_{10m})$ and $S(D)$) are described in Gong (2003) as:

$$W(u_{10m}) = u_{10}^{3.41}$$

$$S(D) = (1 + 0.057r^{1.05}) \times 10^{1.19e^{-B^2}}$$

Where B = (0.380-log r)/0.650. The Emi2 in GEOS uses the size distribution of Gong (2003), wind forcing term proportional to $u_*^{2.41}$ (where u_* is the friction velocity). The default sea salt emissions scheme Emi3 in GEOS further accounts for a SST correction term derived from AOD over the oceans (Randles et al., 2017).

The $T(SST)$ in Emi3 is

T(SST) =

(-1.107211 – 0.010681*tskin_c – 0.002276*tskin_c**2 + 60.288927*1.0/(40.0 – tskin_c))

Here tskin_c is the sea surface skin temperature in Celsius. tskin_c is set to be -0.1 when below -0.1 and to be 36.0 when above 36.0. Furthermore, the overall temperature modification fsstemis is confined within 0.0 to 7. Note that the Emi3 is the default emission currently used in GEOS.

Figure S1 shows the point-to-point comparison of sea salt between ATom PALMS measurements and the GEOS model simulation using the three described emissions along aircraft flight tracks of ATom1 and ATom2, respectively. Several order shifts of sea salt magnitude shown by both measurement and simulation reflect the presence of a strong sea salt vertical gradient within the troposphere. All three emission methods give higher sea salt mass concentrations where the peak values occurred, implying possible overestimation of emissions. Statistical analysis given in table S1 indicates that sea salt simulated by GEOS, over all sampling points of PALMS, is about 1.2 to 2 times higher in ATom1 and 2 to 3 times higher in ATom2 depending on which emission algorithm is used.

To have an accurate sea salt emission, the most important thing is to have a good wind parameterization since wind is the fundamental driver to generate sea spray particles. By using surface friction velocity (Emi2) instead of the 10-m wind (Emi1), the correlation coefficient was increased from 0.50 to 0.54 in ATom1 and from 0.38 to 0.43 in ATom2. This makes sense since surface friction velocity is more physically meaningful than the 10-m wind for sea salt wind-borne emissions.

The correlation coefficients are further lifted up to 0.60 (ATom1) and 0.50 (ATom2) when SST-correction is applied (Emi3, the default emission). Several previous studies have reported that simulated sea salt emission is affected by sea surface temperature. Spada et al. (2013) run an online CTM model to examine five sea salt emission algorithms and found that SST-dependent emission schemes lead to a clear improvement of surface sea salt simulation when compared with measurements of Aerosol Robotic Network (AERONET), University of Miami's Ocean Aerosol Network, and two NOAA Pacific Marine Environmental Laboratory (PMEL) cruises. Salter et al., (2015) proposed a size-resolved particle algorithm and found that total number density decrease nonlinearly with increasing seawater temperature, but other sea salt properties (e.g. effective radius, surface area, volume and mass) increase with increasing seawater temperature due to increased production of particles with dry diameters greater than 1 μm. Jaeglé et al. (2011) used cruise observations to derive an empirical temperature corrected sea salt source function in GEOS-Chem, which resulted in better agreement between simulation and measurements from in situ cruises, MODIS and AERONET AOD. A contradictory conclusion, i.e. no apparent relationship between water temperature and measured sea-salt concentration, however, was found in the analyzed data set, which contained open-ocean shipboard measurements from five different campaigns covering the South Indian Ocean, the Western Pacific region, the New England region, and the Gulf of Maine by the PMEL group (Witek et al., 2007). Overall, inclusion of SST-correction indeed improves sea salt simulation on a global scale at least during the summer and winter seasons. Furthermore, the three emission algorithms discussed in supplementary section show that the uncertainty among the model simulations is generally less than the difference between model and measurement.

Figure Captions

Figure S1a. Sea salt mass concentration with particle diameter (Dp) less than 3 μm along ATom1 flight track. Black line is for PALMS measurement, while color lines represent the GEOS model simulation with three different emission algorithms. Note Emi3 is currently default emission algorithm used in the GEOS model.

Figure S1b. similar to Fig. 2a but for ATom2.

Table S1. Statistical analysis of the sea salt results from the ATom PALMS and SAGA measurements and GEOS5 simulation along flight tracks in ATom1 and ATom2

	Emission algorithm	GEOS – PALMS		
		R	Bias	NRMS
ATom1	Emi1	0.50	1.21	0.11
	Emi2	0.54	2.06	0.15
	Emi3	0.60	2.01	0.15
ATom2	Emi1	0.38	2.00	0.14
	Emi2	0.43	3.09	0.19
	Emi3	0.50	2.55	0.15

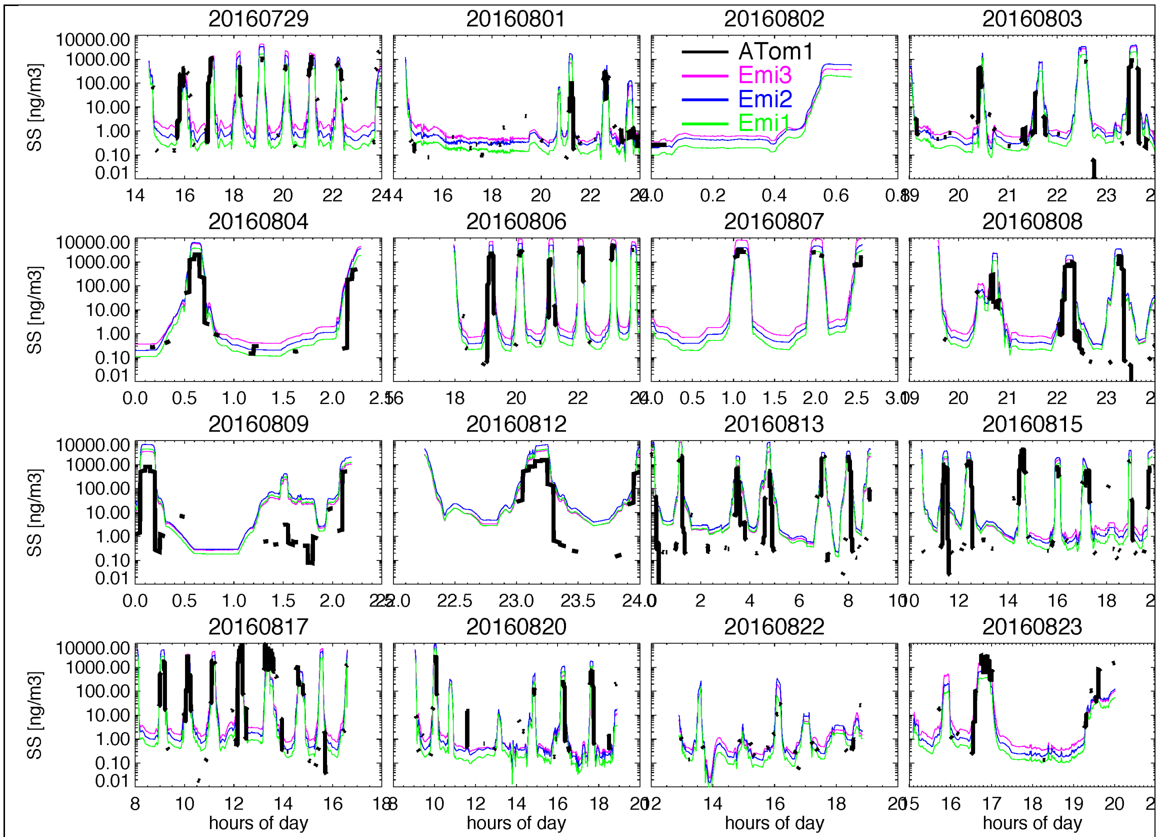

Figure S1a. Sea salt mass concentration with particle diameter (Dp) less than 3 μm along ATom1 flight track. Black line is for PALMS measurement, while color lines represent the GEOS5 model simulation with three different emission algorithms. Note Emi3 is currently default emission algorithm used in the GEOS5 model.

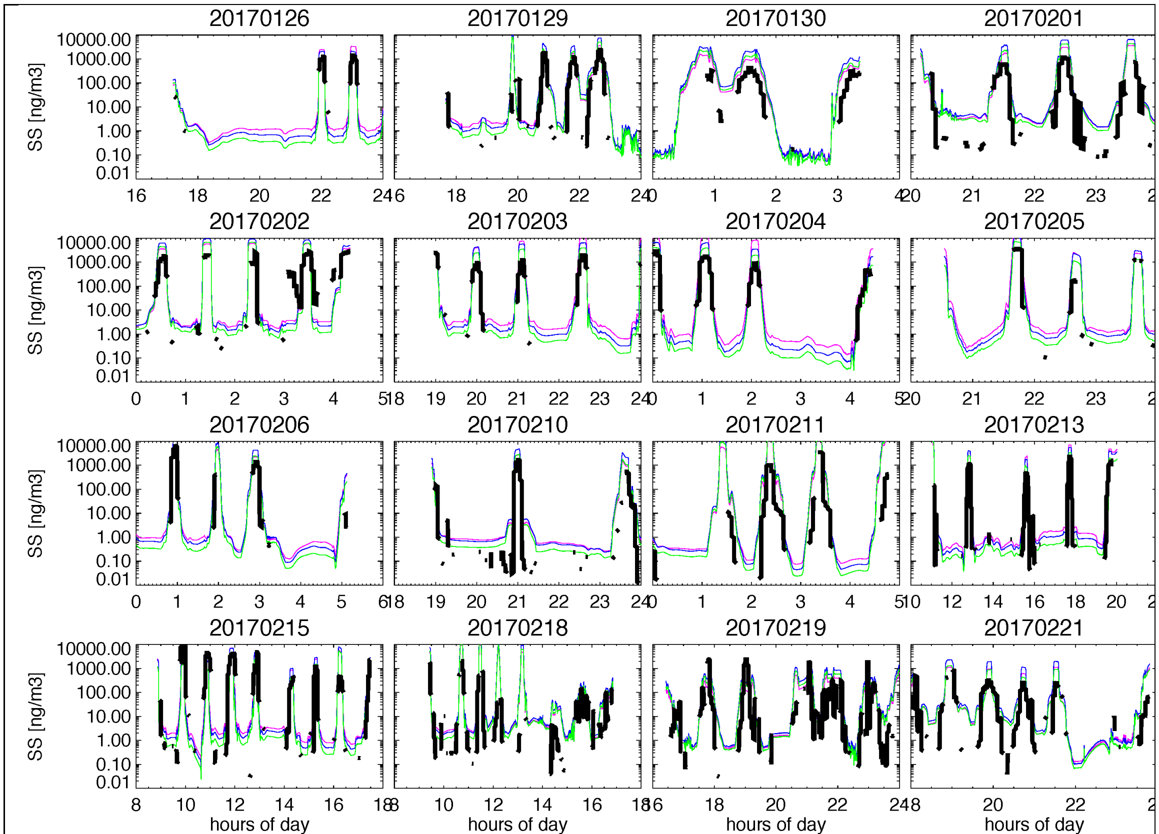

Figure S1b. similar to Figure S1a but for ATom2.