



*Supplement of*

**Seasonal characteristics of emission, distribution, and radiative effect of marine organic aerosols over the western Pacific Ocean: an investigation with a coupled regional climate aerosol model**

**Jiawei Li et al.**

*Correspondence to:* Zhiwei Han (hzw@mail.iap.ac.cn)

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## 1. Marine primary organic aerosol (MPOA) emission

The emission rate of MPOA is the product of sea spray aerosol (SSA) emission rate ( $E_{SSA}$ ) and organic matter fraction of sea spray aerosol ( $OM_{SSA}$ , unitless in a range of 0~1), i.e.

$$E_{MPOA} = \alpha \times E_{SS} \times OM_{SS} \quad (1),$$

where  $OM_{SSA}$  is expressed as a function of wind speed, surface seawater Chl-a concentration, and aerosol size, and  $\alpha$  is a tuning factor. The calculation of  $OM_{SSA}$  follows the method of Gantt et al. (2012):

$$OM_{SS} = \frac{\left( \frac{1}{1 + \exp(X(-2.63[Chl-a]) + X(0.18U_{10}))} \right)}{1 + 0.03 \exp(6.81D_p)} + \frac{0.03}{1 + \exp(X(-2.63[Chl-a]) + X(0.18U_{10}))} \quad (2),$$

where  $U_{10}$  is wind speed at 10 meter ( $m s^{-1}$ ) simulated online by RIEMS-Chem,  $D_p$  is the diameter of sea salt aerosol, and Chl-a is the surface seawater chlorophyll-a concentration ( $mg m^{-3}$ ). The Level-3 daily mean Chl-a concentration retrievals with 9 km resolution from the VIIRS onboard the SNPP satellite platform are used for model inputs and it can reflect day-to-day variation of sea surface Chl-a concentration associated with phytoplankton bloom in the western Pacific.  $X$  is a unitless adjustable coefficient and is set to 3 based on Gantt et al. (2012).

For the tuning factor  $\alpha$ , Gantt et al. (2012) suggested a factor of 6 was able to minimize the relative model biases for the global model GEOS-Chem at two oceanic sites (Mace Head in North Atlantic and Amsterdam Island in remote south Indian Ocean). In this study, we found that a factor of 2 was optimal to obtain the least bias between model simulation and observation over the western Pacific.

## 2. Marine isoprene emission

The sea-air flux of marine isoprene ( $E_{isop}$  in the unit of  $\mu g m^{-2} s^{-1}$ ) is parameterized following the method of Palmer and Shaw (2005), which can be expressed as:

$$E_{isop} = k \times SW_{isop} \quad (3),$$

where  $k$  is the sea-air exchange coefficient ( $cm h^{-1}$ ) and is calculated as:

$$k = 0.31 \times U_{10} \times (660/Sc)^{1/2} \quad (4),$$

where  $Sc$  is the Schmit number of Isoprene.

The surface seawater isoprene concentration  $SW_{isop}$  ( $\mu\text{g m}^{-3}$ ) related to phytoplankton activities is parameterized by the scheme of Gantt et al. (2009):

$$SW_{isop} = H_{\max} \times [Chl-a] \times \int_0^{H_{\max}} EF \ln(I)^2 dh \quad (5),$$

where  $EF$  is the emission factor of isoprene released by phytoplankton,  $I$  is the ambient photosynthetically active radiation (PAR, in the unit of  $\mu\text{Em}^{-2} \text{s}^{-1}$ ),  $H_{\max}$  is the total water depth which isoprene production can occur from the surface to the point and is calculated as:

$$H_{\max} = -\ln\left(\frac{2.5}{I_0}\right) \frac{1}{k_{490}} \quad (6),$$

where  $I_0$  is the all-sky surface incoming solar radiation ( $\text{W m}^{-2}$ ) provided by the model during simulation.  $I_0$  and  $I$  have an approximate relationship of  $1 \text{ W m}^{-2} \approx 2 \mu\text{Em}^{-2} \text{s}^{-1}$ . The diffuse attenuation coefficient values at 490 nm  $k_{490}$  ( $\text{m}^{-1}$ ) is also obtained from VIIRS satellite. The isoprene production is assumed to occur when the light level is greater than  $2.5 \text{ W m}^{-2}$  in surface sea water.

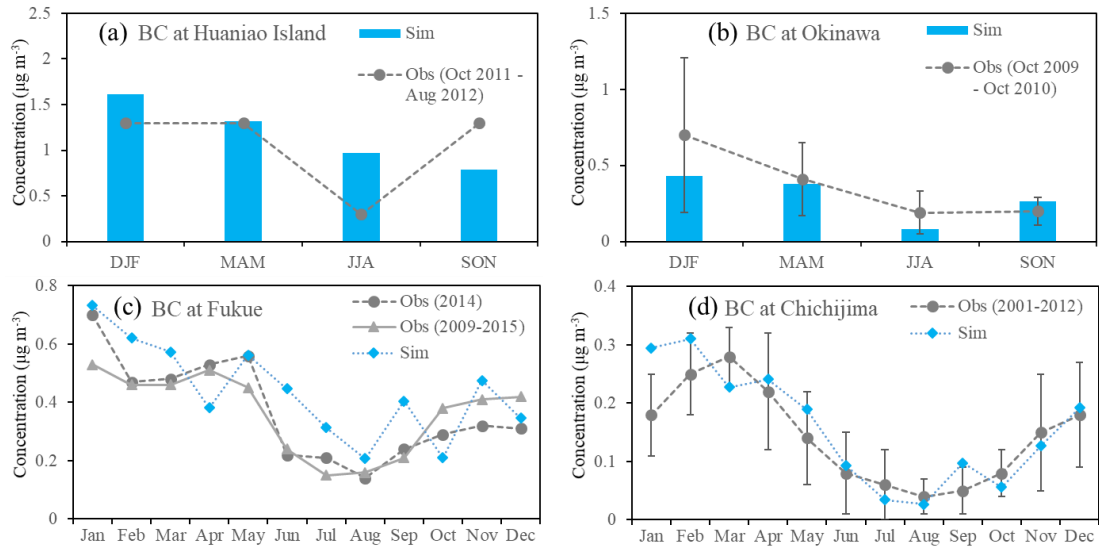


Figure S1. The model simulated (bars) and observed (dotted lines) BC concentrations at different sites. Seasonal mean concentrations were provided at (a) Huaniao Island (Wang et al., 2015) and (b) Okinawa (Kunwar and Kawamura, 2014) while monthly mean concentrations were provided at (c) Fukue (Kanaya et al., 2016) and (d) Chichijima Island (Boreddy et al., 2018). The observed sample standard deviations were available at Okinawa and Chichijima. The simulation is for the year 2014.

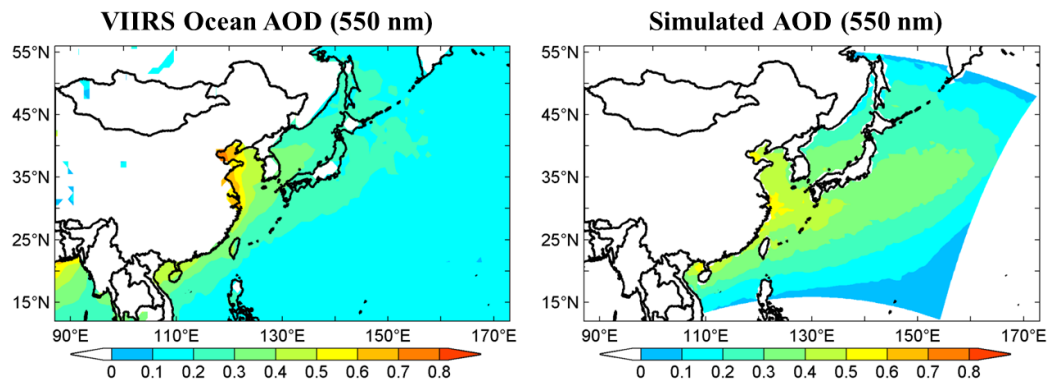


Figure S2. VIIRS retrieved (a) and model simulated (b) annual mean AOD at 550 nm over ocean. Values over the land have been masked. The model results were sampled accordingly to the satellite retrievals.

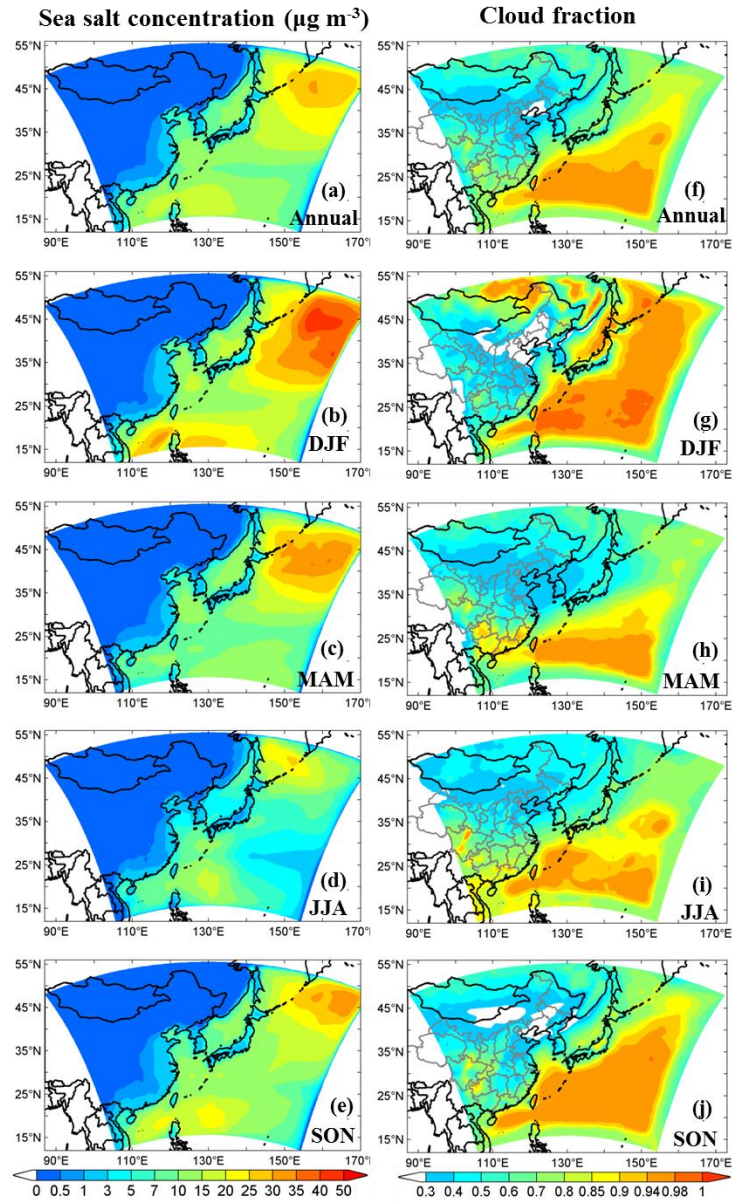


Figure S3. Model simulated annual and seasonal mean near surface sea salt concentrations (unit:  $\mu\text{g m}^{-3}$ ) (a~e) and cloud fractions (unit: %) (f~j).

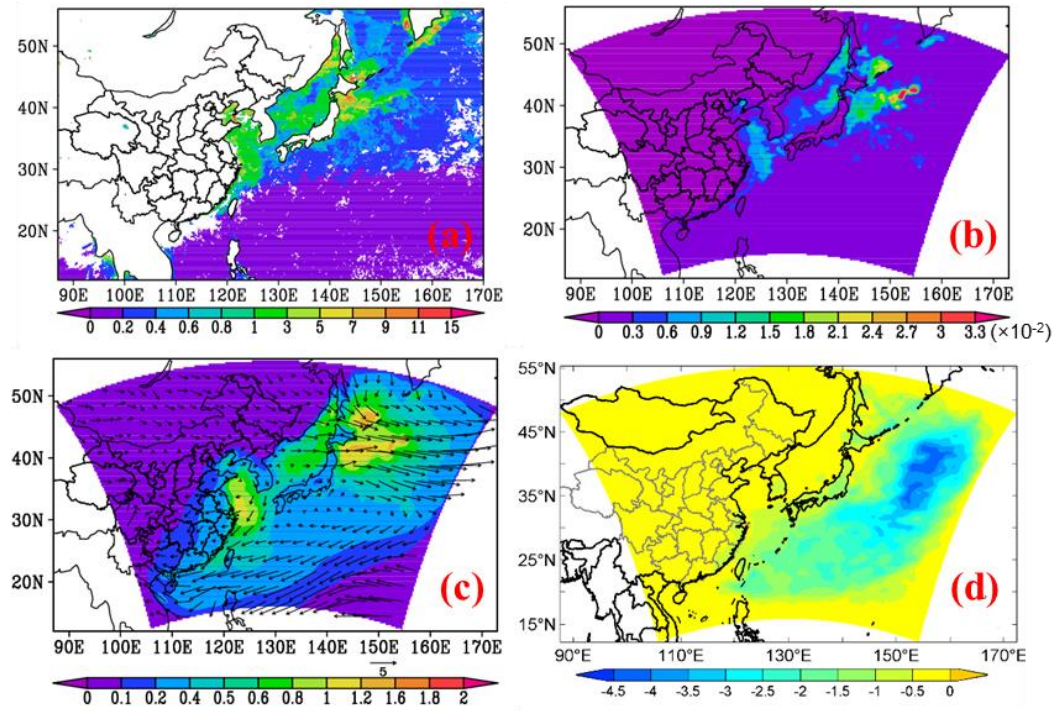


Figure S4. (a) VIIRS retrieved April mean Chl-a concentration (unit:  $\text{mg m}^{-3}$ ), (b) model simulated April mean MPOA emission flux (unit:  $\mu\text{g m}^{-2} \text{s}^{-1}$ ), (c) MOA concentration (unit:  $\mu\text{g m}^{-3}$ ) overlaid with wind vector (unit:  $\text{m s}^{-1}$ ), and (d)  $\text{IRE}_{\text{MOA}}$  (unit:  $\text{W m}^{-2}$ ).

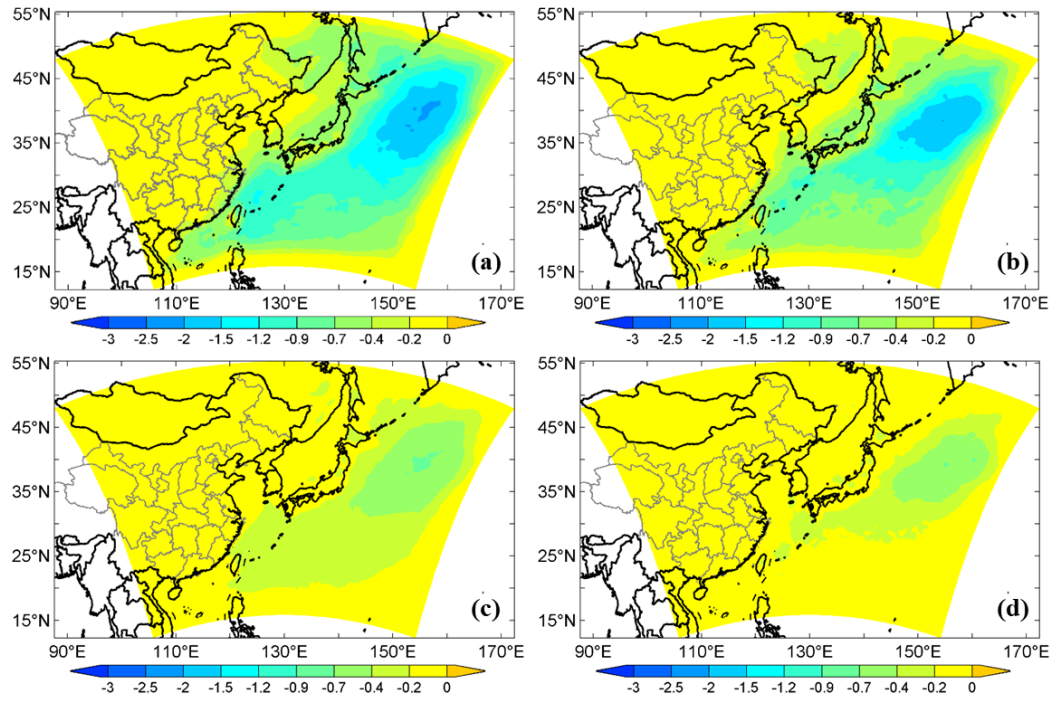


Figure S5. Annual mean IRE due to MOA from different sensitivity simulations. (a) FULL, (b) SENS1, (c) SENS2, and (d) SENS3.

Table S1. Observed (Obs) and simulated (Sim) annual mean sodium (Na<sup>+</sup>) concentrations at EANET sites (units:  $\mu\text{g m}^{-3}$ ). Pearson correlation coefficients (R) are presented.

Site	Samples	Obs	Sim	R
Rishiri	13	2.1	3.4	0.75
Tappi	22	2.9	1.8	0.78
Sado-seki	26	2.3	3.8	0.64
Oki	26	3.7	2.3	0.81
Hedo	24	4.8	2.9	0.53
Ogasawara	26	2.5	2.7	0.85
Total	137	3.1	2.8	0.50



Table S2. Performance statistics for hourly gas precursor concentrations (unit: pptv) at EANET sites for the year 2014. Mean observation (Obs), mean simulation (Sim), correlation coefficient (R), and normalized mean bias (NMB in %) are listed.

Sites	SO <sub>2</sub>				NO <sub>x</sub> /NO <sub>2</sub> <sup>a</sup>				O <sub>3</sub>						
	Samples	Obs	Sim	R	NMB	Samples	Obs	Sim	R	NMB	Samples	Obs	Sim	R	NMB
Rishiri	8493	0.22	0.21	0.62	-5	7606	0.7	0.96	0.27	38	8583	38.6	45.4	0.57	18
Tappi	8570	0.48	0.40	0.35	-15	8517	1.5	2.0	0.40	33	8569	39.3	48.0	0.52	22
Sado	8233	0.38	0.52	0.41	39	8200	0.75	2.9	0.39	286	8400	46.5	49.6	0.55	7
Okii	7837	0.54	0.45	0.54	-17	7919	1.3	1.7	0.39	26	8507	46.8	50.8	0.52	9
Hedo	7546	0.26	0.26	0.55	-2	7910	0.74	1.1	0.47	48	8260	39.6	41.5	0.84	5
Ogasawara	7635	0.10	0.11	0.09	5	7226	0.37	0.37	0.21	1	8506	33.4	35.8	0.84	7
Jeju	8282	0.54	0.76	0.41	42	8300	3.3	3.5	0.29	7	8419	44.7	52.5	0.56	17
Kanghwa	8517	2.77	4.09	0.25	48	8539	5.9	13.0	0.45	120	8526	50.3	29.4	0.55	-42
Imsil	8337	2.32	1.50	0.16	-35	8200	4.0	1.8	0.18	-55	8385	31.4	39.9	0.54	27
Average	73450	0.86	0.95	0.51	10	72417	2.1	2.9	0.48	36	76155	41.2	43.6	0.54	6

a: NO<sub>x</sub> in Japan and NO<sub>2</sub> in Korea.

Table S3. Model simulations from this study and cruise observation-based estimates from previous studies for marine isoprene emission fluxes over different areas of the western Pacific of East Asia (Units: nmol m<sup>-2</sup> day<sup>-1</sup>).

Regions of cruises <sup>a</sup>	Observations		References	Simulations (this study)	
	periods	mean (max/min)		periods	mean (max/min)
Western North Pacific (30.75~35.78°N, 146.42°E)	18–26 May 2001	140.0 (300.0/32.0)	Matsumaga et al. (2002)	May 2014	85.0 (100.0/40.0)
Northwest Pacific (34~43°N, 138~150°E)	Jan 2008–2012	21.4	Ooki et al. (2015)	Jan 2014	26.0
	May 2008–2012	143.8		May 2014	89.0
	Aug 2008–2012	55.6		Aug 2014	63.0
East China Sea (25.69~30°N, 121~126°E)	Oct–Nov 2013	48.3 (169.2/4.2)	Li et al. (2018)	Oct–Nov 2013	48.0 (120.0/0)
	May–Jun 2014	36.1 (137.8/2.5)		May–Jun 2014	35.0 (180.0/0)
South Yellow Sea and East China Sea (30~38°N, 121.6~127°E)	14 July – 1 Aug 2013	161.5 (537.2/22.17)	Li et al. (2017)	Jul 2014	130.0 (240.0/0)

a: Longitude and latitude ranges indicate the coverages of each cruise.

Table S4. Modeled domain and annual/seasonal mean IRE due to MOA from different sensitivity simulations.

	WP <sup>a</sup>	ECS <sup>b</sup>	NWP <sup>c</sup>
	ANN		
FULL	-0.66	-0.23	-1.0
SENS1	-0.53	-0.13	-0.81
SENS2	-0.20	-0.06	-0.34
SENS3	-0.14	-0.03	-0.24

a: Mean over oceanic areas.

b: 27~40°N, 115~123°E.

c: 35~55°N, 140~160°E.

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