

Atmospheric Organosulfate Formation Regulated by Continental Outflows and Marine Emissions over East Asian Marginal Seas

Shubin Li¹, Yujue Wang^{1, 2, *}, Yiwen Zhang¹, Yizhe Yi¹, Yuchen Wang⁴, Yuqi Guo¹, Chao Yu¹, Yue Jiang¹, Jinhui Shi^{1, 2}, Chao Zhang^{1, 2}, Jialei Zhu⁵, Wei Hu⁵, Jianzhen Yu^{6, 7}, Xiaohong Yao^{1, 2}, Huiwang Gao^{1, 2}, Min Hu^{3, *}

¹Frontiers Science Center for Deep Ocean Multispheres and Earth System, Key Laboratory of Marine Environment and Ecology, Ministry of Education of China, Ocean University of China, Qingdao, China

²Laboratory for Marine Ecology and Environmental Science, Qingdao Marine Science and Technology Center, Qingdao, China

³State Key Joint Laboratory of Regional Environment and Sustainability, College of Environmental Sciences and Engineering, Peking University, Beijing 100871, China

⁴College of Environmental Science and Engineering, Hunan University, Changsha, Hunan, 410082, China

⁵Institute of Surface-Earth System Science, School of Earth System Science, Tianjin University, Tianjin, China

⁶Division of Environment & Sustainability, Hong Kong University of Science & Technology, Hong Kong, China

⁷Department of Chemistry, Hong Kong University of Science & Technology, Hong Kong, China

*Correspondence to: Yujue Wang (wangyujue@ouc.edu.cn); Min Hu (minhu@pku.edu.cn)

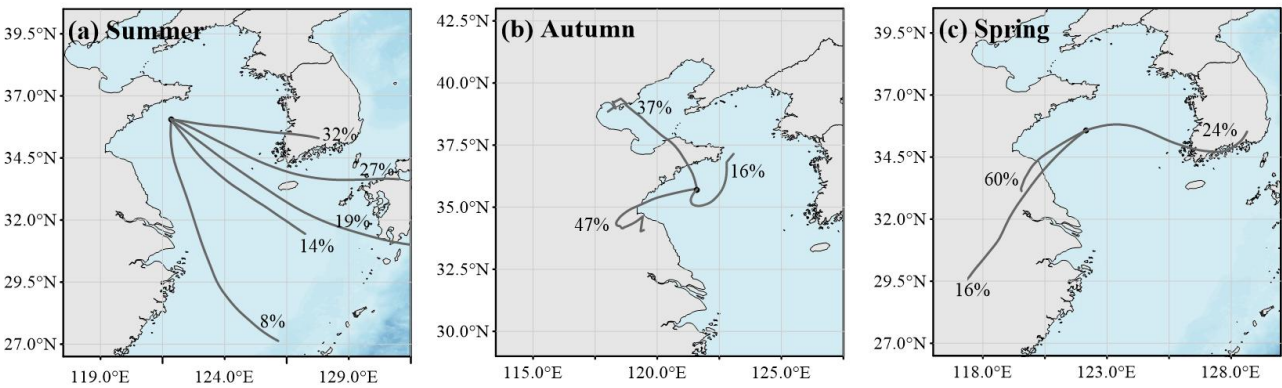
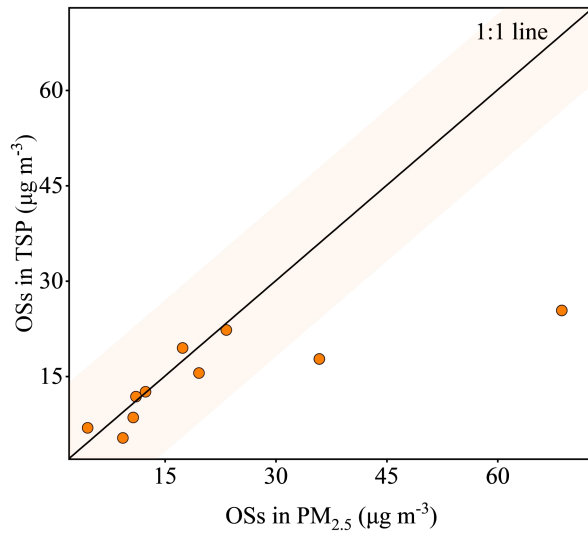


Figure S1 The 72h back trajectories of air masses over the YBS during the observation in (a) summer (15 July–26 July), (b) autumn (21 Oct.–2 Nov.), and (c) spring (14 April–25 April). Trajectories at the center site of the observation region were calculated during each cruise: 36.1°N, 122. 0°E for the summer 2021 and spring 2022 cruises, and 35.7°N, 121.7°E for the autumn 2021 cruise.



25 **Figure S2** Comparison of OSs mass concentrations in the $\text{PM}_{2.5}$ and the TSP samples during the autumn cruise over YBS.

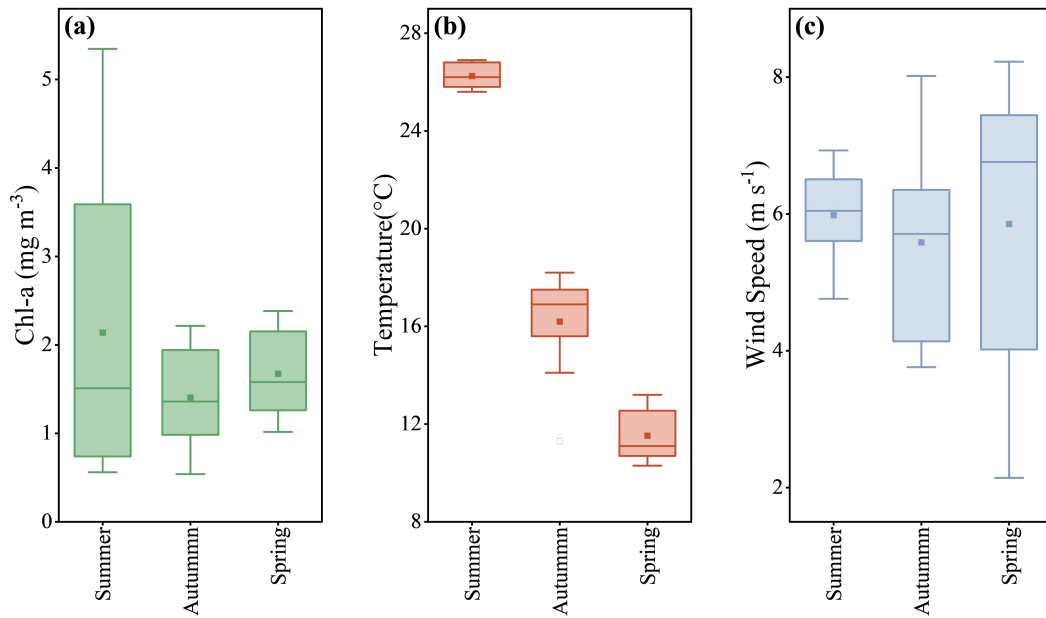


Figure S3 Seasonal variations of (a) *Chl-a*, (b) temperature, and (c) wind speed during the observations. The markers represent the mean values and the boxes represent the ranges of 25th–50th–75th percentiles.

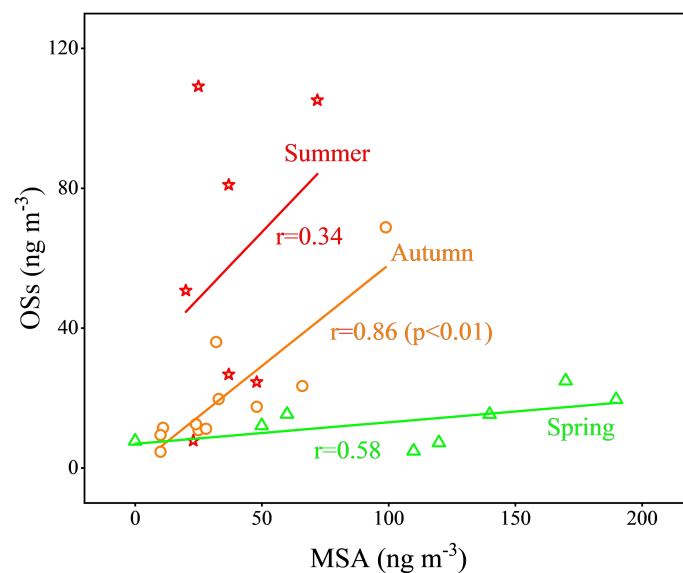


Figure S4 Correlations of OSs and MSA in marine aerosols in summer (red), autumn (yellow), and spring (green).

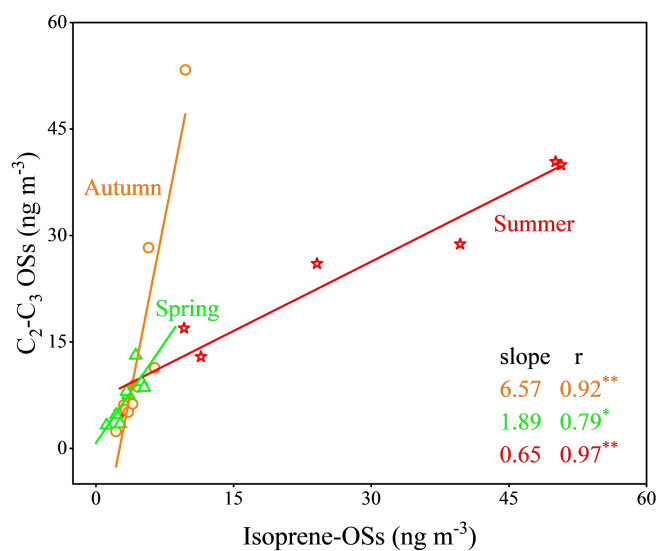
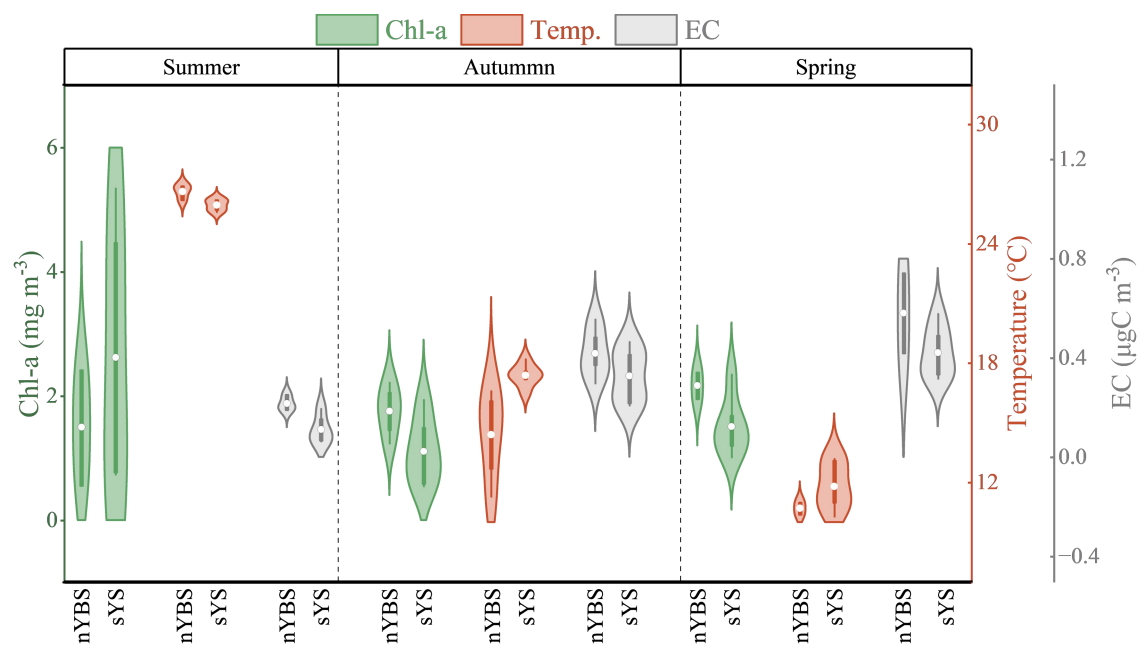


Figure S5 Correlations of C₂-C₃ OSs and isoprene-OSs in marine aerosols over the YBS during summer (red), autumn (yellow), and spring (green). The values listed in the panel are the curve slopes (mass ratios) of C₂-C₃ OSs versus isoprene-OSs and their correlation coefficients (* p<=0.05, ** p<=0.01).



40 **Figure S6** Comparison of chlorophyll-a (*Chl-a*) in surface seawater, air temperature, and particulate EC concentrations over the nYBS and sYS during the summer, autumn, and spring cruises.

Table S1 Atmospheric OSs and NOSs quantified in this study and their concentrations during each cruise over the YBS.

Common name	Formula	[M-H] ⁻	Standard	Concentrations (ng/m ³)		
				Summer (n=7)	Autumn (n=11)	Spring (n=8)
Hydroxyacetone sulfate (HAS)	C ₃ H ₅ O ₅ S ⁻	152.99	HAS	2.13±1.25	1.28±0.39	0.98±0.28
Glycolic acid sulfate (GAS)	C ₂ H ₃ O ₆ S ⁻	154.97	GAS	20.69±10.41	10.01±13.43	5.05±2.72
Lactic acid sulfate (LAS)	C ₃ H ₅ O ₆ S ⁻	168.98	LAS	1.40±0.83	1.54±0.77	1.12±0.38
ΣC ₂ -C ₃ OSs				24.22±12.44	12.84±14.38	7.15±3.13
Methacrylic acid epoxide (MAE)-OS	C ₄ H ₇ O ₇ S ⁻	198.99	LAS	2.33±1.66	1.06±0.42	0.46±0.24
Isoprene-OS (m/z 211)	C ₅ H ₇ O ₇ S ⁻	210.99	LAS	11.15±7.44	2.71±1.35	2.19±0.92
Isoprene epoxydiol (IEPOX)-OS	C ₅ H ₁₁ O ₇ S ⁻	215.02	LAS	10.64±7.63	0.49±0.19	0.52±0.17
Isoprene NOSs	C ₅ H ₁₀ NO ₉ S ⁻	260.01	LAS	2.82±2.09	0.31±0.16	0.29±0.16
ΣIsoprene OSs/NOSs				26.94±2.09	4.57±2.01	3.46±1.31
Monoterpene OSs	C ₁₀ H ₁₇ O ₅ S ⁻	249.08	α-Pinene OS β-Pinene OS limonene OS	0.02±0.02	0.03±0.01	0.02±0.01
	C ₉ H ₁₅ O ₆ S ⁻	251.06	limonaketone OS	0.96±0.99	0.49±0.35	0.26±0.12
Monoterpene NOSs	C ₁₀ H ₁₆ NO ₇ S ⁻	294.06	α-Pinene NOS limonene NOS	5.45±6.39	2.24±2.48	2.18±3.18
	C ₉ H ₁₄ NO ₈ S ⁻	296.04	limonaketone NOS	0.20±0.21	0.22±0.19	0.19±0.16
ΣMonoterpene OSs/NOSs				6.63±7.60	2.98±2.74	2.65±3.39
Total quantified OSs/NOSs				57.79±38.93	20.39±19.73	13.25±8.34

Table S2 Principal Component Analysis of the measured compounds in marine aerosols over the YBS.

Compounds	PC1 (52%)	PC2 (21%)	PC3 (10%)
HAS	0.93	0.13	0.07
GAS	0.81	0.48	0.15
LAS	0.59	0.70	0.21
MAE-OS	0.96	0.10	-0.15
C ₅ H ₇ O ₇ S ⁻	0.97	-0.07	0.02
IEPOX-OS	0.96	-0.22	-0.02
Isoprene-NOS	0.96	-0.17	0.07
C ₁₀ H ₁₇ O ₅ S ⁻	0.42	0.38	-0.06
Limonaketone-OS	0.86	0.23	-0.03
C ₁₀ H ₁₆ NO ₇ S ⁻	0.69	-0.04	0.18
C ₉ H ₁₄ NO ₈ S ⁻	0.32	0.19	0.12
MSA	-0.11	0.08	0.83
EC	-0.38	0.39	0.51
nss-K ⁺	-0.13	0.95	0.01
NO ₃ ⁻	-0.06	0.95	0.15
nss-SO ₄ ²⁻	0.95	0.05	-0.11
Cl ⁻	0.53	0.45	0.64
Na ⁺	0.07	0.01	0.87