

**Atmospheric deposition promotes relative abundance of
main dimethylsulfoniopropionate producers in the western
North Pacific**

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Method S1. Water sampling, nutrient and aerosol amendments and incubation.

To avoid vessel and other metal contamination as much as possible, we sailed out a wooden boat (Huaniao) or reached the HDPE water pipe out about 3 meters from the gunwale tied on a fishing rod, with a ceramic plumb hanging on the end (cruise). On the island, we filled up the 50 L carboys using plastic buckets and funnels, while during the cruise, the air compressor (Jiebao, 8BAR) and diaphragm pump (Wilden, P100/PVDF+Teflon, USA) were introduced to pump water up. All sampled water was covered with black bags and filtered through 150 μ m nylon mesh tied to the faucet of carboys before experiments, removing larger zooplankton. After dividing into PP cubitainers (Paerl et al., 1990) or HDPE culture bottles and adding nutrient solutions or aerosol extracts, these marked treatments were soaked in the seawater (Huaniao: tied on the ropes fixed by fish rafts; cruise: floated in a cistern equipped with cyclic ballast seawater) and covered with a gray screen to weaken the sunlight intensity, simulating the light condition in the surface layer. In the laboratory, the illumination time and temperature were controlled to mimic the change of day and night. All the apparatuses used in the experiments, including pipes, carboys, cubitainers, measuring cylinder, etc. were dipped and washed with 0.2 M ultrapure HCl and then rinsed twice with MilliQ water in the laboratory before; and finally rinsed by sampled seawater when dividing initial water.

$(\text{NH}_4)_2\text{SO}_4$ (NH_4^+), NaNO_3 (NO_3^-), KH_2PO_4 (PO_4^{3-}) and Cu/Fe standard solution (ShijiAoke Biotechnology co. LTD, Beijing, China) were used for preparing the stock solutions of nutrients. Aerosols were previously sampled at Huaniao and analyzed by

Ion Chromatography (ICS3000, DIONEX, for determination of ions) and Inductively Coupled Plasma Optical Emission Spectroscopy (SPECTRO, Germany, for detection of total and soluble elements). Backward trajectories were applied for further analysis (HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model from the NOAA Air Resource Lab). Aerosol membranes were cut into small pieces and ultrasonicated within MilliQ water for 40 min, acquiring the supernate and stored at 4°C. Extracts were usually prepared just before division and addition.

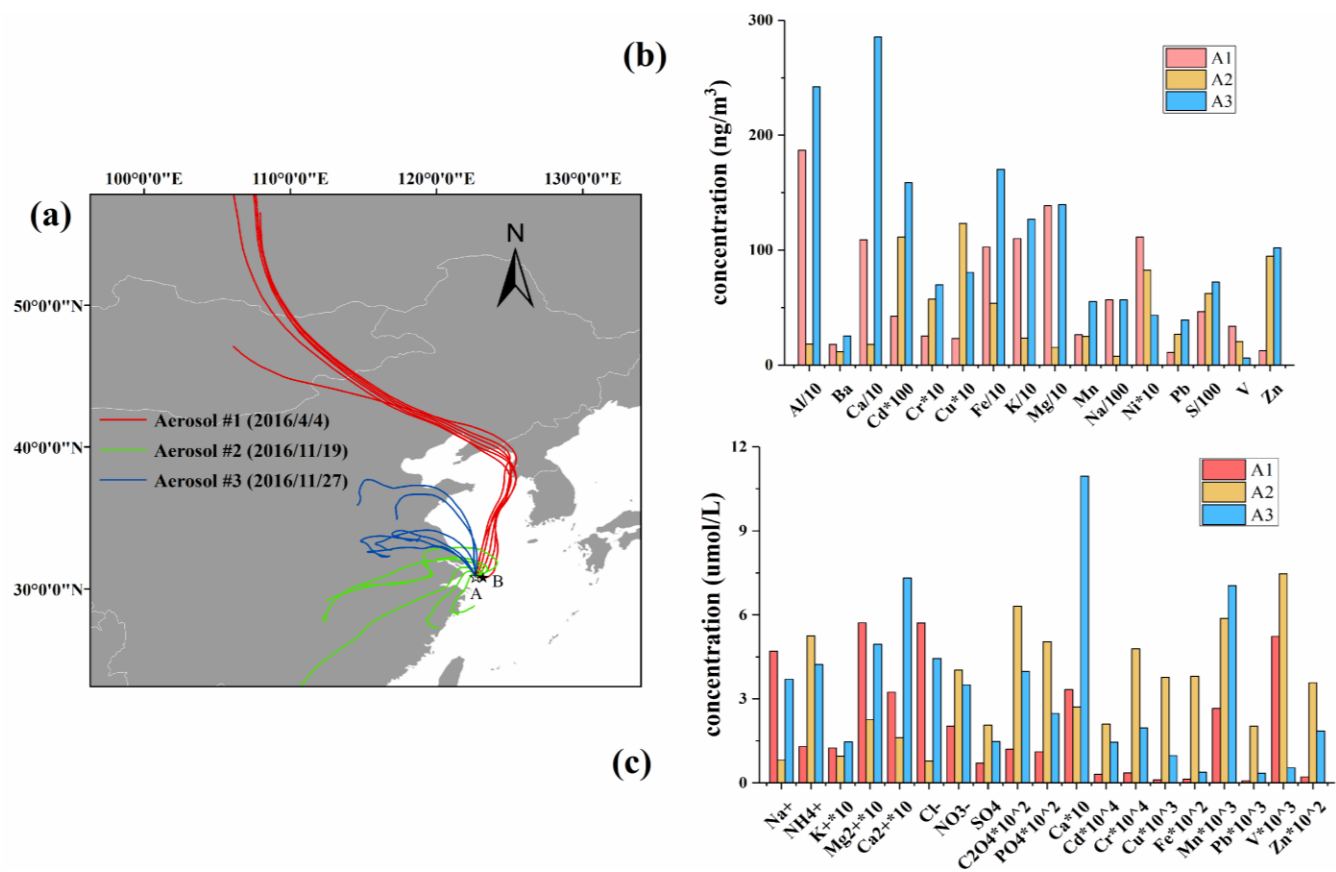


Figure S1-1. The (a) air mass back trajectories; (b) composition; (c) additive concentration of soluble elements of three aerosols used in Huaniao Island (HN). (a-A: aerosol sampling site; a-B: water sampling site)

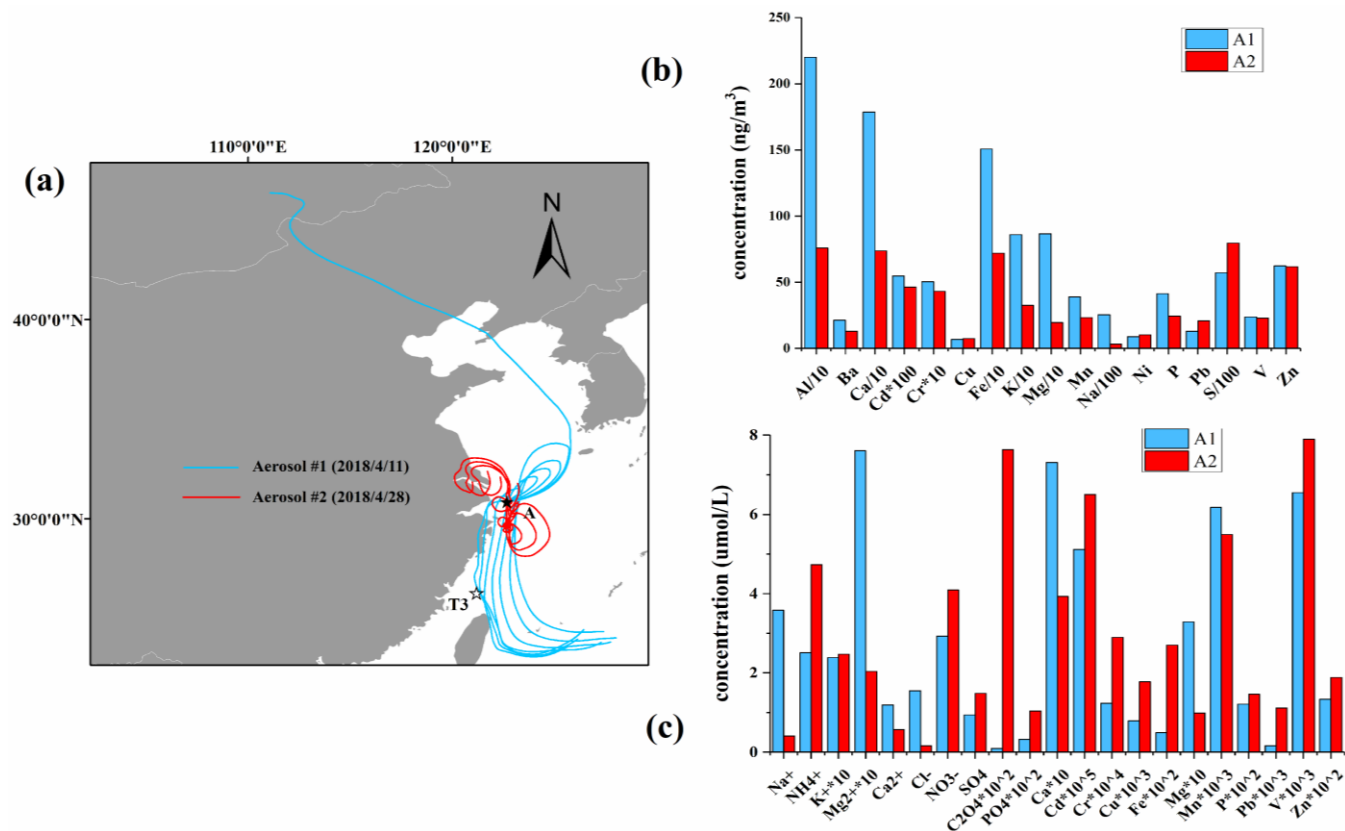


Figure S1-2. The (a) air mass back trajectories; (b) composition; (c) additive concentration of soluble elements of aerosols used in Taiwan Strait (TWS). (a-A: aerosol sampling site; a-T3: water sampling site)

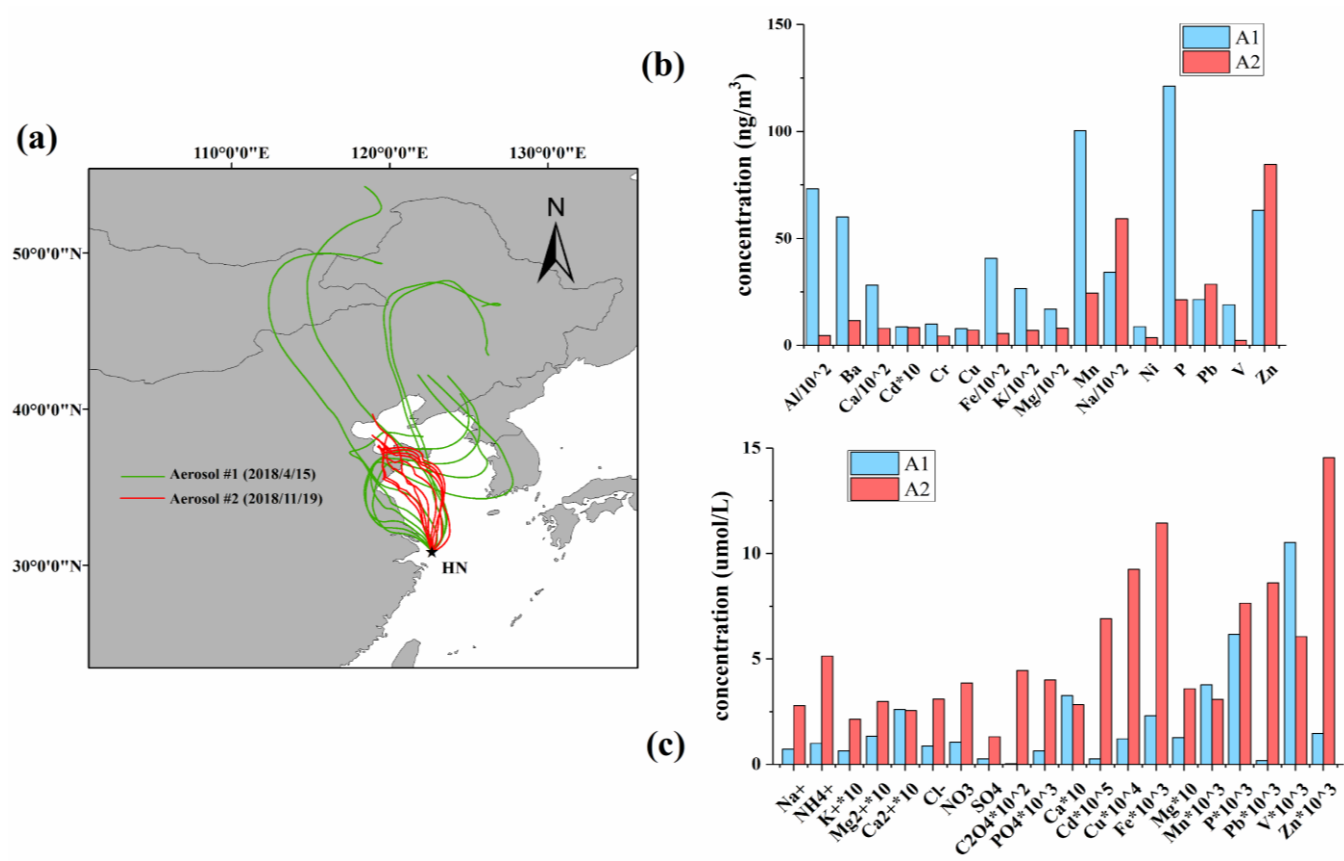


Figure S1-3. The (a) air mass back trajectories; (b) composition; (c) additive concentration of soluble elements of aerosols used in North Pacific Subtropical Gyre (NPSG).

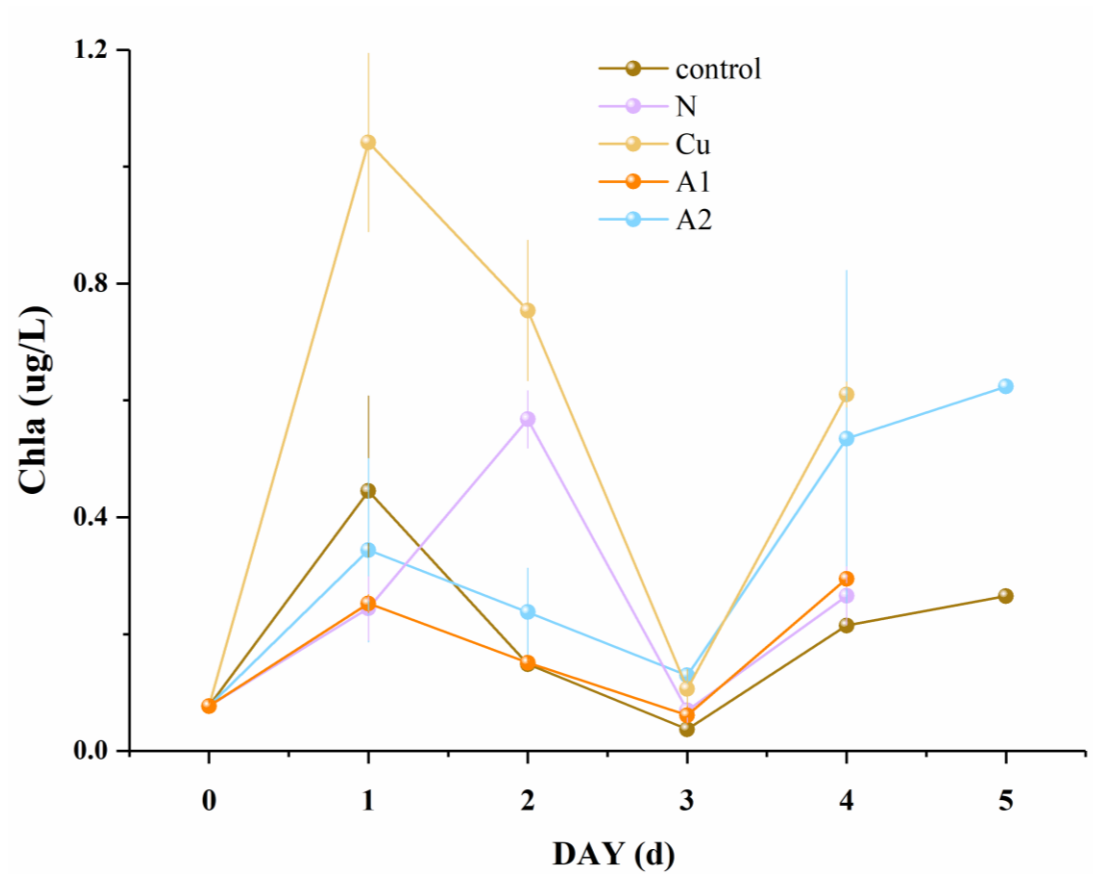


Figure S2. Variation of *Chl a* concentrations in different treatments of the bioassay experiment in Taiwan Strait (TWS).

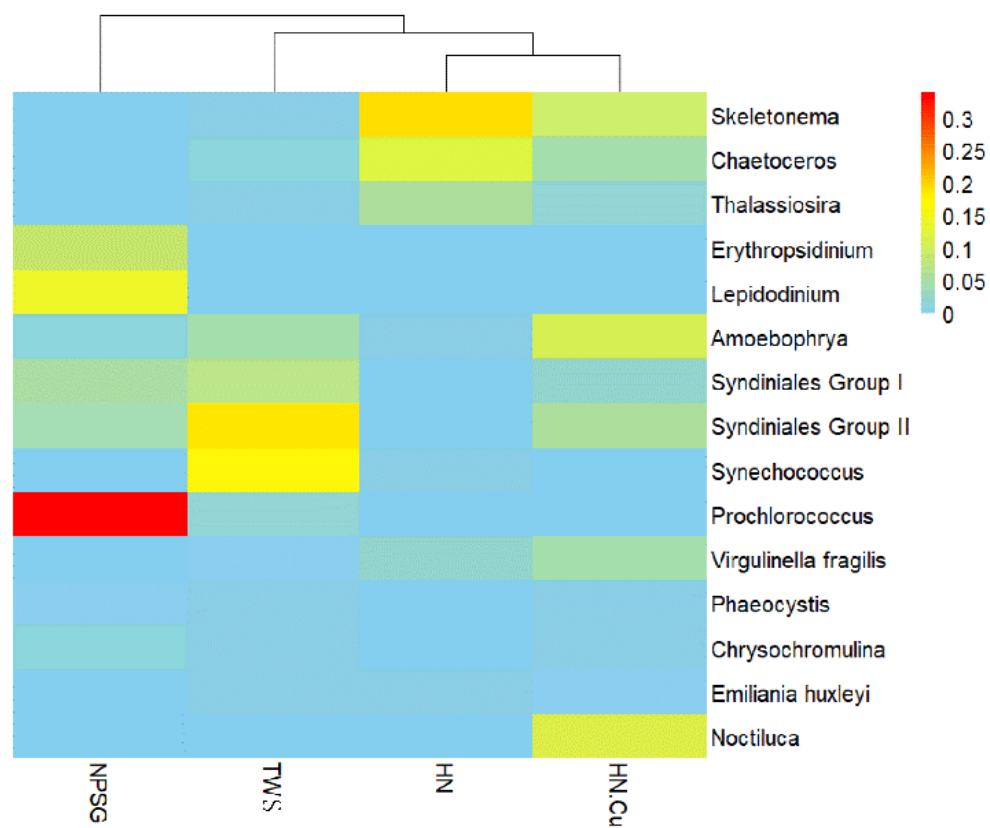


Figure S3. Relative abundance of different taxa on species level in the initial seawaters used for the bioassay experiments at Huaniao lab (HN.Cu), Huaniao Island (HN), Taiwan Strait (TWS) and North Pacific Subtropical Gyre (NPSG).

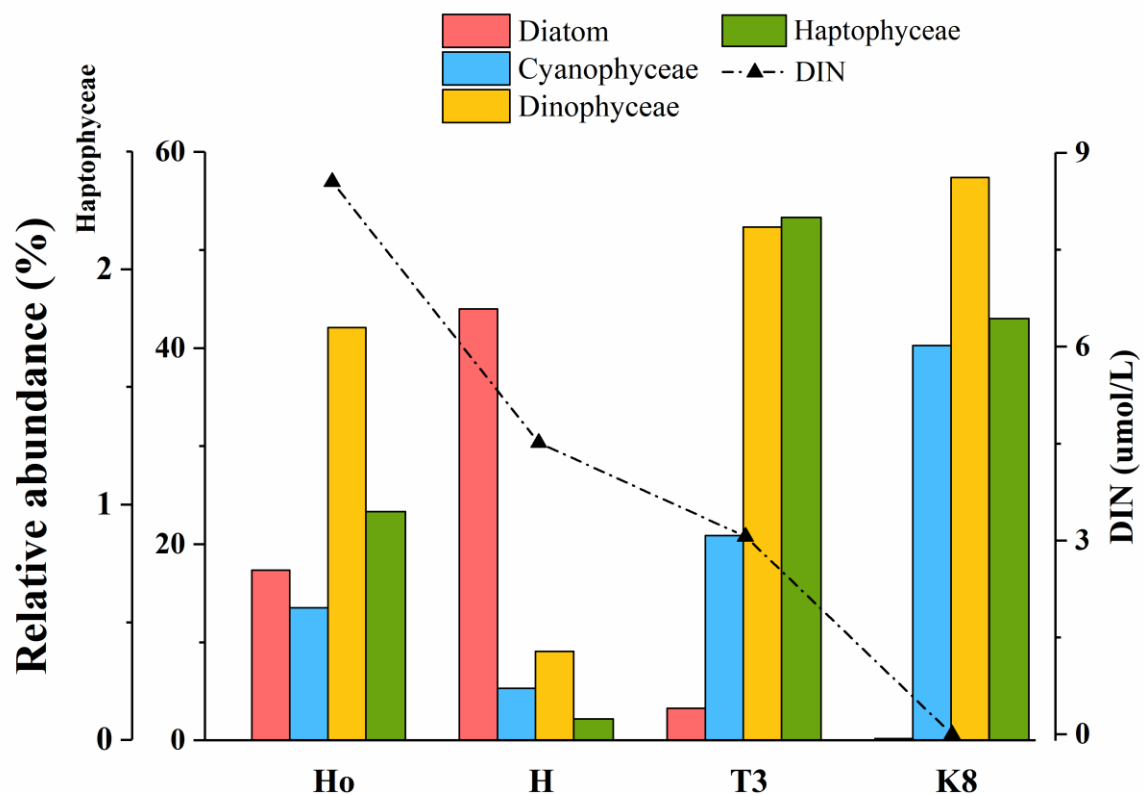


Figure S5. Relative abundances of prime algae classes and DIN concentrations in the initial seawaters sampled from Huaniao Lab (Ho), Huaniao Island (H), Taiwan Strait (T3) and North Pacific Subtropical Gyre (K8).

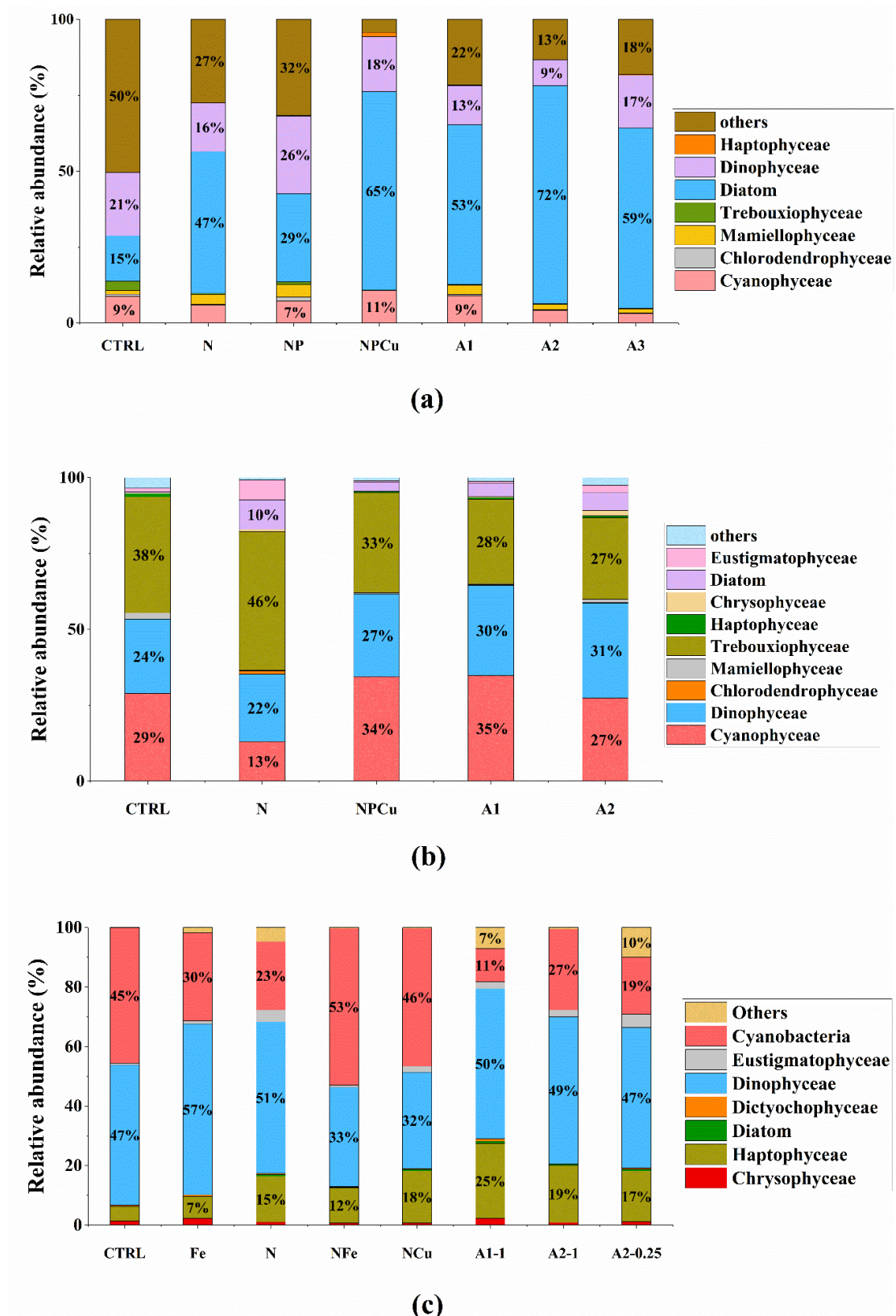


Figure S6. Relative abundances of dominant phytoplankton groups in different treatments during the bioassay experiments at (a) Huaniao Island (HN), (b) Taiwan Strait (TWS), and (c) North Pacific Subtropical Gyre (NPSG).

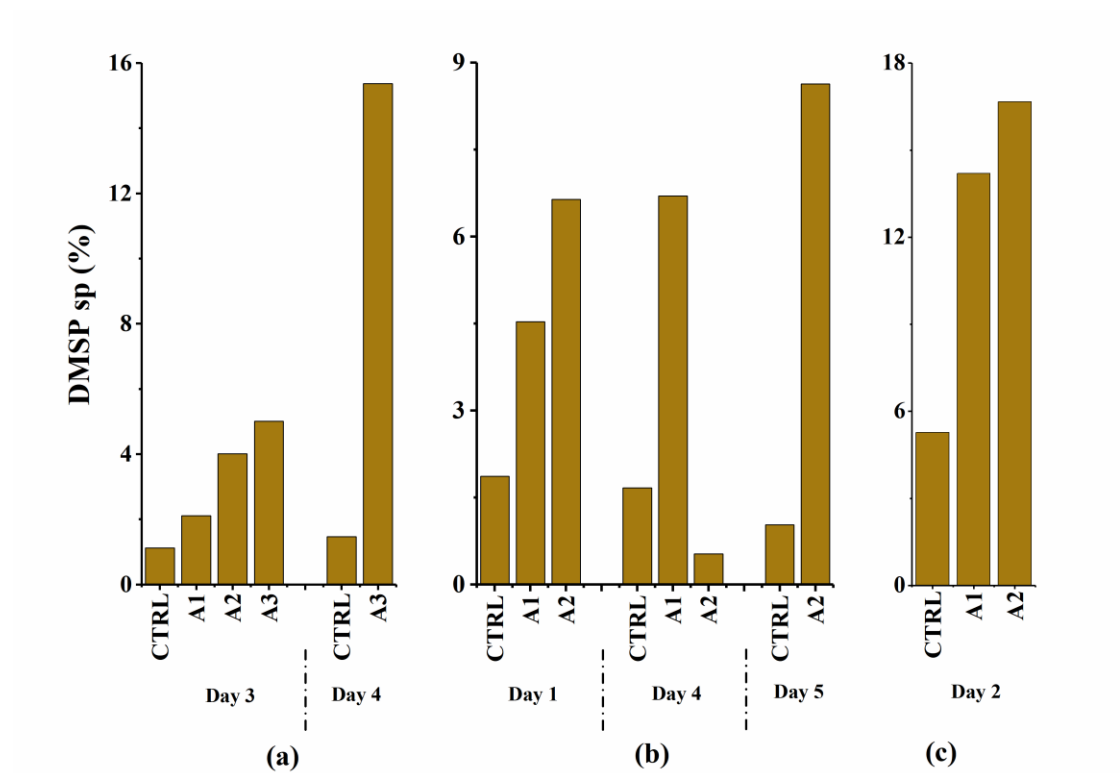


Figure S7. Percentages of DMSP species in the control (CTRL) and aerosol treatments in the late period (after 3 days) of HN (a), in both early and late periods (Day 1, 4 & 5) of TWS (b) and at the maximum *Chl a* (Day 2) of NPSG incubation (c)

Table S1. The release concentrations of DMSP by phytoplankton on cultures (Keller et al., 1989).

class	species	pg DMSP/cell	uM DMSP/cm3 CV
Dinophyceae	Gymnodinium sp.	24	124.63
	<i>Chrysochromulina</i>	3.62	412.69
Haptophyceae	<i>Emiliana huxleyi</i>	0.75	166.42
	Phaeocystis sp.	2.29	260.45

Table S2. Basic information of four bioassay experiments. (all aerosols used were sampled at Huaniao Island)

	date	water sampling	incubation	aerosols
				Aerosol #1 2016.4.4
HN	2018.5.4	30°52'N, 122°40'E; N/P=26.18	<i>in situ</i>	Aerosol #2 2016.11.19
				Aerosol #3 2016.11.27
HN Lab	2019.4.6	30°50'N, 123°10'E; N/P=43.03	laboratory	-
				Aerosol #1 2018.4.11
TWS	2018.7.14	26°17'N, 121°12'E; N/P=18.10	<i>in situ</i>	Aerosol #2 2018.4.28
				Aerosol #1 2018.4.15
NPSG	2019.5.28	11°N, 155°E; DIN BDL *	<i>in situ</i>	Aerosol #2 2018.11.19

* DIN concentration below the detection limit 0.1 umol/L.

Table S3. Relative abundance of high DMSP production associated species (primarily Dinophyceae and Haptophyceae) in different treatments of the bioassay experiments conducted at Huaniao Island (HN), Taiwan Strait (TWS) and North Pacific Subtropical Gyre (NPSG).

	Dinophyceae			Haptophyceae		Total
	<i>Gyrodinium</i>	<i>other Dino</i> *	<i>Phaeocystis</i>	<i>Chrysochromulina</i>	<i>Emiliana huxleyi</i>	
HN-CTRL	0.00%	0.54%	0.03%	0.00%	0.06%	0.63%
HN-N	0.00%	0.53%	0.01%	0.00%	0.01%	0.54%
HN-NP	0.00%	0.89%	0.05%	0.01%	0.06%	1.01%
HN-NPCu	0.28%	1.50%	0.28%	0.07%	0.25%	2.38%
HN-A1	0.00%	0.32%	0.12%	0.00%	0.07%	0.51%
HN-A2	0.00%	0.15%	0.01%	0.00%	0.01%	0.17%
HN-A3	0.00%	0.29%	0.20%	0.00%	0.08%	0.57%
HN-Cu-CTRL	0.03%	0.92%	0.04%	0.37%	0.03%	1.39%
HN-Cu-NFeCu	0.05%	1.24%	0.01%	0.08%	0.01%	1.39%
HN-Cu-10xCu	0.03%	1.47%	0.05%	0.55%	0.05%	2.15%
TWS-CTRL	1.52%	1.87%	0.07%	0.54%	0.00%	4.00%
TWS-N	0.11%	5.21%	0.00%	0.02%	0.04%	5.38%
TWS-NPCu	0.53%	4.09%	0.07%	0.09%	0.11%	4.88%

TWS-A1	0.43%	3.80%	0.10%	0.02%	0.18%	4.53%
TWS-A2	0.28%	5.82%	0.09%	0.14%	0.32%	6.64%
NPSG-CTRL	2.16%	0.67%	0.39%	2.06%	0.00%	5.27%
NPSG-Fe	3.31%	1.01%	0.66%	1.78%	0.02%	6.78%
NPSG-N	6.91%	1.30%	2.60%	4.90%	0.05%	15.76%
NPSG-NFe	3.52%	0.40%	1.53%	4.52%	0.00%	9.98%
NPSG-NCu	2.97%	0.28%	2.09%	6.97%	0.00%	12.32%
NPSG-A1-1	1.03%	1.88%	3.54%	7.64%	0.12%	14.20%
NPSG-A2-1	5.64%	1.12%	2.23%	7.68%	0.00%	16.67%
NPSG-A2-0.25	2.58%	1.69%	2.36%	5.34%	0.05%	12.02%

* *other Dino* contains *Symbiodinium* sp., *Alexandrium minutum*, *Prorocentrum* sp., *Gyrodinium impudicum*, *Scrippsiella*, *Dinophysis acuminata*, *Heterocapsa pygmaea*, *Amphidinium carterae*, *Cryptothecodinium cohnii*, *Heterocapsa triquetra*, *Gymnodinium nelson*, *Alexandrium tamarense*, *Thoracosphaera heimii*, *Gymnodinium* sp., *Gonyaulax spinifera*, etc. (Caruana and Malin, 2014)

Appended Table. Relative abundance of high DMSP production associated species (class) in the late period of HN incubation.

	CTRL	A1	A2	A3
Haptophyceae	24.17%	61.89%	24.69%	15.37%
Dinophyceae	72.41%	20.16%	62.61%	81.16%
other	3.43%	17.96%	12.70%	3.47%

Table S4. Concentration of important total elements and ions and classification of aerosols used in bioassay experiments.

possible sources			HN			TWS		NPSG	
			A1	A2	A3	A1	A2	A1	A2
total elements (ng/m3)	dust	Al	1869.595	181.662	2419.583	2199.673	758.496	7325.567	456.758
		Ca	1090.186	178.837	2854.493	1786.467	737.002	2811.971	794.180
		Fe	1027.774	536.818	1701.107	1507.556	719.439	4073.124	550.022
		Mn	26.433	25.036	55.304	39.020	23.129	100.394	24.474
	vehicles	Cd	0.425	1.113	1.587	0.547	0.464	0.862	0.834
		Cu	2.311	12.336	8.061	6.670	7.331	7.843	7.095
		Zn	12.614	94.727	101.897	62.278	61.702	63.144	84.500
	industry	Pb	10.964	26.636	39.265	12.819	20.789	21.390	28.559
	vessels	V	33.816	20.394	6.170	23.678	22.924	18.988	2.284
	ions (ug/m3)	Na ⁺	10.589	0.872	5.437	3.658	0.721	3.262	5.542
		Mg ²⁺	1.342	0.254	0.760	0.810	0.374	0.628	0.619
		Cl ⁻	19.848	1.304	10.079	4.885	0.442	6.052	9.510
	anthropogenic	NH ₄ ⁺	2.285	4.448	4.859	4.014	6.524	3.510	8.000

	NO ₃ ⁻	12.269	11.740	13.846	16.128	19.414	12.817	20.733
	SO ₄ ²⁻	6.628	9.298	9.051	8.001	10.905	5.019	10.917
	C ₂ O ₄ ²⁻	0.103	0.261	0.224	0.007	0.514	0.005	0.339
				soil dust (urban) +				marine +
classification		dust + marine	urban pollution	anthropogenic	dust + marine	urban pollution	dust	anthropogenic
				pollutants				pollutants

Table S5. Diversity index (Shannon & Simpson index) of different treatments.

Treatments	shannon	simpson
HN-CTRL	4.8233	0.9325
HN-N	4.3215	0.8995
HN-NP	5.4179	0.9559
HN-NPCu	3.7827	0.7937
HN-A1	3.8864	0.8348
HN-A2	4.4521	0.8466
HN-A3	4.3712	0.8846
HN-Cu-CTRL	3.7863	0.8310
HN-Cu-NFeCu	3.5111	0.8029
HN-Cu-10xCu	4.5050	0.9073
TWS-CTRL	3.7223	0.7931
TWS-N	3.6339	0.7732
TWS-NPCu	3.7053	0.8035
TWS-A1	4.0108	0.8354
TWS-A2	4.4927	0.8728
NPSG-CTRL	5.2044	0.9308
NPSG-Fe	4.8032	0.8763
NPSG-N	5.6090	0.9518
NPSG-NFe	4.9064	0.9167
NPSG-NCu	5.3510	0.9543
NPSG-A1-1	5.9617	0.9744
NPSG-A2-1	5.6555	0.9663
NPSG-A2-0.25	5.8608	0.9673

Table S6. Relative abundance of DMSP species in aerosol treatments in the early and late period of incubation in HN and TWS.

	early	DMSP species	late	DMSP species
HN	A1-D1	0.51%	A1-D3	2.11%
	A2-D1	0.17%	A2-D3	4.01%
	A3-D1	0.57%	A3-D3	5.01%
			A3-D4	15.36%
TWS	A1-D1	4.53%	A1-D4	6.70%
	A2-D1	6.64%	A2-D5	8.63%

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