Supplementary Materials

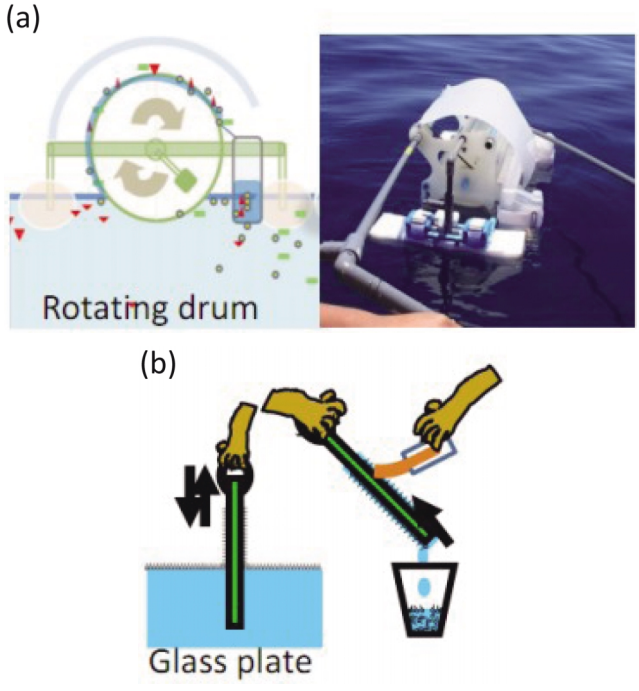
Introduction

This supplement contains two figures and details of the SML sampling devices and of the procedures for glass-plate, rotating-drum during the current study and two tables.

SML sampling method and procedure

The glass plate sampler (surface area of 0.18 m^2) was vertically immersed into seawater and withdrawn with an approximate speed of 20 cm/s (Harvey and Burzell, 1972), and adhesive surface seawater retained on the glass plate was wiped with a polypropylene wiper and collected into polypropylene bottles pre-washed with acid. The in-house PMMA rotating drum sampler with surface area of 0.79 m^2 (diameter of 250 mm) was operated manually with an approximate rotating speed of 6 rpm, and adhesive seawater retained on the drum was wiped out with a polypropylene wiper into collection bottles.

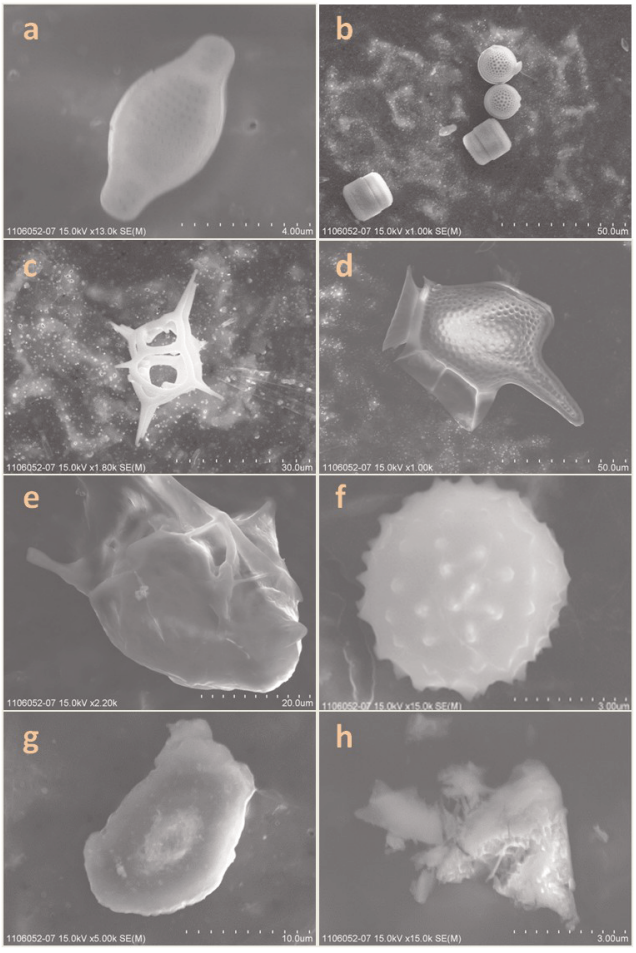
Just before sampling, devices were kept covered with a cleaned polypropylene sheet to prevent contamination by atmospheric deposition or dust; at every sampling, the first samples were used to clean and rinse the devices and were then discarded. After sampling, all parts of the sampling devices in contact with sampling water were specially rinsed thoroughly with Milli-Q water, oven dried and kept in a clean polypropylene box to prevent excess particles and dust, while the rest of the sampling devices and other related equipment were rinsed vigorously with freshwater, air dried and kept dry and covered with a clean sheet until the next sampling.



Supplementary Fig. S1. Devices and the operating procedures used for sampling the sea-surface microlayer: a) rotating drum and b) glass plate.

Reference

Harvey, G. W. and Burzell, L. A. (1972) A simple microlayer method for small samples. *Limnol. Oceanogr.* **17**(1), 156–157, doi:10.4319/lo.1972.17.1.0156.



Supplementary Fig. S2. Micrographs showing the particles classified as (a–b) diatoms, (c) silico-flagellates, (d) dino-flagellates, (e–f) other microorganisms, (g) other particles, organic-like and (h) other particles, inorganic-like (Fe-containing particles).

Supplementary Table S1. Sampling locations, SML thicknesses, meteorological conditions, and the concentrations of dissolved phosphorus (μM), particulate phosphorus (nM), and elemental compounds (nM) (Al, Si, S, and Ca and Fe)

Station description				Method	Thickness	Chl-a	Dissolved phosphorus		Particulate phosphorus		Particulate element conc.				
					(μm)	μg/l	SRP (μM)	%DOP	TPP (nM)	%POP	pAl (μM)	pSi (μM)	pS (μM)	pCa (μM)	pFe (nM)
Station 01	0°N, 95.5°W	Water (°C)	23.1	SML-GP	45.70	—	0.84 ± 0.14 (1.0)	n.a.	587.8 ± 129.9 (32.7)	58.59					
Date	Feb. 02, 2012	Air (°C)	25.9	SML-D	38.10	—	0.85 ± 0.30 (1.0)	n.a.	1286.2 ± 43.5 (71.5)	57.42	0.14 ± 0.04 (2.8)	2.94 ± 0.05 (6.1)	1.83 ± 0.03 (6.3)	0.54 ± 0.05 (3.9)	81.0 ± 13.0 (11.3)
Time	08:34–09:36	WS (m/s)	2.9	SUR*	0.30	0.2	0.70 ± 0.03	n.a.	15.4 ± 2.7	48.82	L.D.	0.20 ± 0.04	0.14 ± 0.02	0.06 ± 0.01	22.3 ± 4.30
(LT = UTC-7)				SSW*	1.50	—	0.88 ± 0.09	n.a.	18.0 ± 11.90	64.44	0.05 ± 0.01	0.48 ± 0.06	0.29 ± 0.01	0.14 ± 0.01	7.2 ± 2.20
Station 02	0°N, 100°W	Water (°C)	24.5	SML-GP	45.60	—	0.45 ± 0.02 (1.3)	58.72	9.6 ± 2.5 (2.6)	29.41					
Date	Feb. 03, 2012	Air (°C)	26.5	SML-D	35.80	—	0.57 ± 0.16 (1.6)	47.71	12.9 ± 10.1 (3.5)	27.45	0.11 ± 0.02 (n.a.)	1.08 ± 0.05 (5.7)	0.14 ± 0.02 (1.2)	0.14 ± 0.01 (2.3)	25.0 ± 2.2 (5.2)
Time	10:36–11:52	WS (m/s)	2.4	SUR*	0.30	0.2	0.54 ± 0.12	50.46	13.5 ± 2.7	60.45	L.D.	0.23 ± 0.07	0.12 ± 0.01	0.38 ± 0.07	2.3 ± 2.20
(LT = UTC-7)				SSW*	1.50	—	0.35 ± 0.06	59.77	3.7 ± 3.3	25.55	L.D.	0.19 ± 0.02	0.12 ± 0.01	0.06 ± 0.01	4.8 ± 2.20
Station 05	0°N, 115°W	Water (°C)	24.7	SML-GP	32.20	—	0.97 ± 0.05 (0.9)	45.51	25.8 ± 14.7 (2.8)	69.03					
Date	Feb. 07, 2012	Air (°C)	25.9	SML-D	33.40	—	1.14 ± 0.03 (1.0)	44.93	41.6 ± 2.0 (4.5)	74.85	0.11 ± 0.03 (1.0)	3.19 ± 0.03 (2.3)	0.42 ± 0.01 (6.0)	0.16 ± 0.01 (1.8)	42.2 ± 6.9 (2.8)
Time	12:48–13:53	WS (m/s)	1.1	SUR*	0.30	0.2	0.93 ± 0.12	36.73	23.6 ± 2.7	42.71	L.D.	0.70 ± 0.14	0.10 ± 0.03	0.05 ± 0.01	L.D.
(LT = UTC-7)				SSW*	1.50	—	1.14 ± 0.03	46.23	9.3 ± 1.5	31.90	0.11 ± 0.04	1.37 ± 0.11	0.07 ± 0.01	0.09 ± 0.01	15.2 ± 5.80
Station 08	0°N, 130°W	Water (°C)	24.1	SML-GP	48.30	—	0.95 ± 0.04 (0.9)	49.74	15.6 ± 13.5 (1.6)	57.71					
Date	Feb. 11, 2012	Air (°C)	25.1	SML-D	39.60	—	1.10 ± 0.08 (1.1)	53.19	10.9 ± 6.6 (1.1)	58.89	L.D.	0.63 ± 0.02 (3.3)	0.32 ± 0.02 (8.0)	0.13 ± 0.01 (2.6)	45.4 ± 10.7 (5.0)
Time	09:23–10:58	WS (m/s)	2.7	SUR*	0.30	0.2	1.10 ± 0.08	55.82	13.5 ± 2.0	70.22	L.D.	0.29 ± 0.04	0.08 ± 0.02	0.04 ± 0.01	4.8 ± 4.30
(LT = UTC-8)				SSW*	1.50	—	1.03 ± 0.15	48.76	9.5 ± 3.2	71.25	L.D.	0.19 ± 0.02	0.04 ± 0.01	0.05 ± 0.01	9.0 ± 6.90
Station 10	0°N, 140°W	Water (°C)	23.6	SML-GP	55.60	—	0.57 ± 0.02 (1.1)	56.82	25.7 ± 15.8 (1.7)	74.44					
Date	Feb. 13, 2012	Air℃(C)	24.8	SML-D	41.40	—	0.66 ± 0.03 (1.3)	42.61	30.2 ± 24.3 (2.0)	61.80	0.11 ± 0.03 (1.6)	2.56 ± 0.24 (1.0)	0.39 ± 0.04 (3.5)	0.24 ± 0.02 (4.0)	38.1 ± 15.7 (n.a.)
Time	09:00–10:20	WS (m/s)	4.4	SUR*	0.30	0.4	0.50 ± 0.03	52.83	17.3 ± 2.0	65.27	0.06 ± 0.04	2.24 ± 0.12	0.13 ± 0.01	0.04 ± 0.01	L.D.
(LT = UTC-9)				SSW*	1.50	—	0.52 ± 0.06	46.94	14.8 ± 2.0	78.86	0.07 ± 0.01	2.58 ± 0.16	0.11 ± 0.01	0.06 ± 0.01	L.D.

WS = Wind speed 30 min. average.
*Method; SML-GP = Glass plate, D = Drum; *Depth in meters.*
n.a. = Data not available; L.D. = Less than limit of detection; DL_{Al} = 0.04 μM; DL_{Fe} = 2 nM.
Number in parentheses indicates the Enrichment Factor, unit of the particulate Fe is nM.

Supplementary Table S2. The average relative percentages of elemental composition of classified particle types characterized by SEM/EDX single-particle analysis for particles in the SML and SSW collected at station 1. The sum of the raw EDX signal intensities of all elements in the table is treated as 100% for each analyzed particle (examined 3–5 times for each particle) and averaged with the total numbers of analyzed particles in the same group. If the elemental signal did not exceed S/N = 3, the element was treated as "not detected".

Particle group	No. particles		Relative percentages of elemental composition												
	Total	P_{detect}	Na	Mg	Al	Si	P	S	Cl	K	Ca	Fe	Cu	Zn	Ti
SSW particles	45	5					[1.8]								
Diatoms	18	3	20.2	4.9	—	46.8	0.1	3.65	21.9	—	0.4	—	—	—	—
Other microorganisms	8	2	29.5	11.3	—	6.9	4.2	6.5	42.7	0.2	1.1	—	1.2	—	—
Other particles	19	0													
Organic-like	10	0	40.0	3.3	—	—	—	0.95	45.3	0.4	0.1	—	—	—	—
Inorganic-like	9	0	25.3	3.2	9.7	43.3	—	0.59	11.3	4.2	1.6	0.7	—	—	—
Type I-Fe-particle (0.4–2 μm)	(10)		25.9	1.4	5.7	53.7	—	—	3.9	6.0	2.2	1.2	—	—	—
SML particles	66	51					[4.1]								
Diatoms	15	10	11.2	1.4	—	70.2	2.8	1.4	10.3	—	0.1	—	—	—	—
Other microorganisms	24	20	24.2	3.9	0.1	16.7	5.8	10.4	36.6	0.2	1.7	—	1.5	—	—
Other particles	27	21													
Organic-like	6	5	25.3	4.6	—	0.0	3.9	7.9	41.6	—	—	—	—	—	—
Inorganic-like	21	16	12.7	2.8	0.3	9.8	3.6	2.5	20.7	—	0.3	17.5	0.1	—	0.4
Type I-Fe-particle (0.4–2 μm)	(2)		10.0	3.5	0.7	4.0	5.4	0.5	22.5	—	0.7	18.0	—	—	1.3
Type II-Fe-particle (6–10 μm)	(5)		1.8	0.1	0.1	0.4	0.2	0.4	7.9	—	—	34.5	0.4	—	—

Number in [brackets] represents the $\%P_{avg}$.