Supporting information

Air-sea exchange and atmospheric deposition of phthalate esters in the South China Sea

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Content

Table S16. Air-sea exchange fluxes of PAEs in the South China Sea. Positive value indicates water to air volatilization and negative value indicates air to water deposition

Table S17. Particle dry deposition of PAEs in the South China Sea

Text S1. Chemicals and materials

PAEs standard mixture, containing Dimethyl phthalate (DMP), Diethyl phthalate (DEP), Diisobutyl phthalate (DiBP), Di-*n*-butyl phthalate (D*n*BP), Butylbenzyl phthalate (BBP), Dicyclohexyl phthalate (DCHP) and Di(2-ethylhexyl) phthalate (DEHP) at 1000 µg/mL each, and surrogate standards, e.g. deuterated dimethyl phthalate (DMP d4), diethyl phthalate (DEP d4), n-dibutyl phthalate (DnBP d4) and di(2-ethylhexyl) phthalate (DEHP d4), were supplied by LGC (Wesel, Germany).

Picograde® solvents, e.g. methanol, acetone, n-hexane and dichloromethane (DCM) were purchased from Promochem (Germany). Neutral silica gel (0.063-0.2 mm) was supplied by Macherey Nagel (Düren, Germany) and anhydrous sodium sulphate (99%) and XAD-2 resin was obtained from Merck (Darmstadt, Germany).

Text S2. Washout ratio

Considering both vapor and particle scavenging mechanism, washout ratio (W) can be expressed as equation 1,

$$W = (1-ø)(RT/H)+øWp \qquad (1)$$

where ø is the particle-bound fraction of the chemical, RT/H is the dimensionless Henry's law constant (H) at the ambient temperature (298 K), and WP is the particle scavenging coefficient[1]. A representative WP value of 20,000 is used for the calculation in this work.[2]

Text S3. Air-sea exchange

During the transport from continental sources to the South China Sea, PAEs may undergo several processes at the interface between air and seawater. The direction and fluxes of air-sea exchange of PAEs are mainly controlled by their physicochemical parameters such as Henry's Law Constants and the concentrations in air and seawater, and are influenced by metrological conditions. In this work, air-sea exchange fluxes of PAEs were calculated using the modified version of Whitman's two-film fugacity model, which has been extensively applied for the evaluation of PAHs, PCBs and PAEs in the marine environment,[3-5] The volatilization and deposition fluxes (F_{vol} and F_{dep}, ng/m^2/day) are calculated as in equation 3 and 4,

$$K_{OL} = \left(\frac{1}{K_w} + \frac{1}{K_a * H}\right)^{-1} \quad (2)$$

$$F_{vol} = K_{OL} C_w \quad (3)$$

$$F_{dep} = K_{OL} C_a / H' \quad (4)$$

The net diffusive gas exchange flux (F, ng/m^2/day) is then calculated by subtracting the volatilization flux from the deposition flux, which is described as equation 5,

$$F = K_{OL}\left(C_w - \frac{C_a}{H'}\right) \quad (5)$$

where Cw (ng/m^3) and Ca (ng/m^3) are the dissolved- and gas-phase concentrations, (Cw-Ca/H´) describes the concentration gradient (ng/m^3), H´ is the dimensionless Henry's law constant. The K_{OL} (m/s) is the overall mass transfer coefficient, which contains contributions from the mass transfer coefficients of the water phase (kw) and the air phase (ka). The calculation of Kw and Ka for PAEs refer to the equations recommended by Schwarzenbach et al.[6] and Wanninkoff's quadratic relationship.[7] H´ of PAEs are corrected with temperature of seawater (Tw, K) and averaged salt concentration (Cs, 0.5 mol/L). The total propagation error in F was 45%, which is derived from an error propagation analysis from previous studies.[5] A positive F value indicates seawater to air volatilization, while a negative F means that deposition is dominating air-sea exchange flux.

Text S4. Atmospheric particle deposition

Atmospheric deposition is one of the key processes that remove pollutants from air and is considered the major source of organic matter and chemical pollutants into the marine environment. Based on the concentrations of

PAEs measured in the particle phase, the particle-bound dry deposition fluxes (F_D, ng/m^2/day) were calculated via equation (6)[6]:

$$F_D = C_p \times V_d \quad (6)$$

where Cp is the concentrations of particle phase PAEs (ng/m^3), and V_d is the deposition velocity (cm/s), which can be used to estimate the deposition fluxes of air pollutants. In this work, we used the dry deposition velocity of 0.2 cm/s, which was estimated for pollutants concentrated in fine particles over the South China Sea.[8]

Text S5. Pearson correlation analysis

A correlation analysis of PAEs in air and seawater was conducted. Pearson correlation coefficient values of DiBP and DnBP ($p < 0.01$, $r \geq 0.951$), and DMP and DEP ($p < 0.01$, $r \geq 0.647$) (Table S14) indicated significant correlations between them, and can be attributed to similar sources and transport pathways. DEHP did not show any correlation with other PAEs. In seawater, DEP was highly correlated with DMP ($p < 0.01$, $r \geq 0.659$), BBP ($p < 0.01$, $r \geq 0.578$) and DEHP ($p < 0.01$, $r \geq 0.578$) (Table S15), and DEHP had a correlation with most other PAEs, except from DCHP. We suppose that PAEs have uniform sources for the seawater, while PAEs in air might be affected by their original sources, monsoon, and atmospheric degradation and precipitation.

Figure S1. Air sampling stations in the South China Sea, with the blue dots marked at the starting position of each air sample

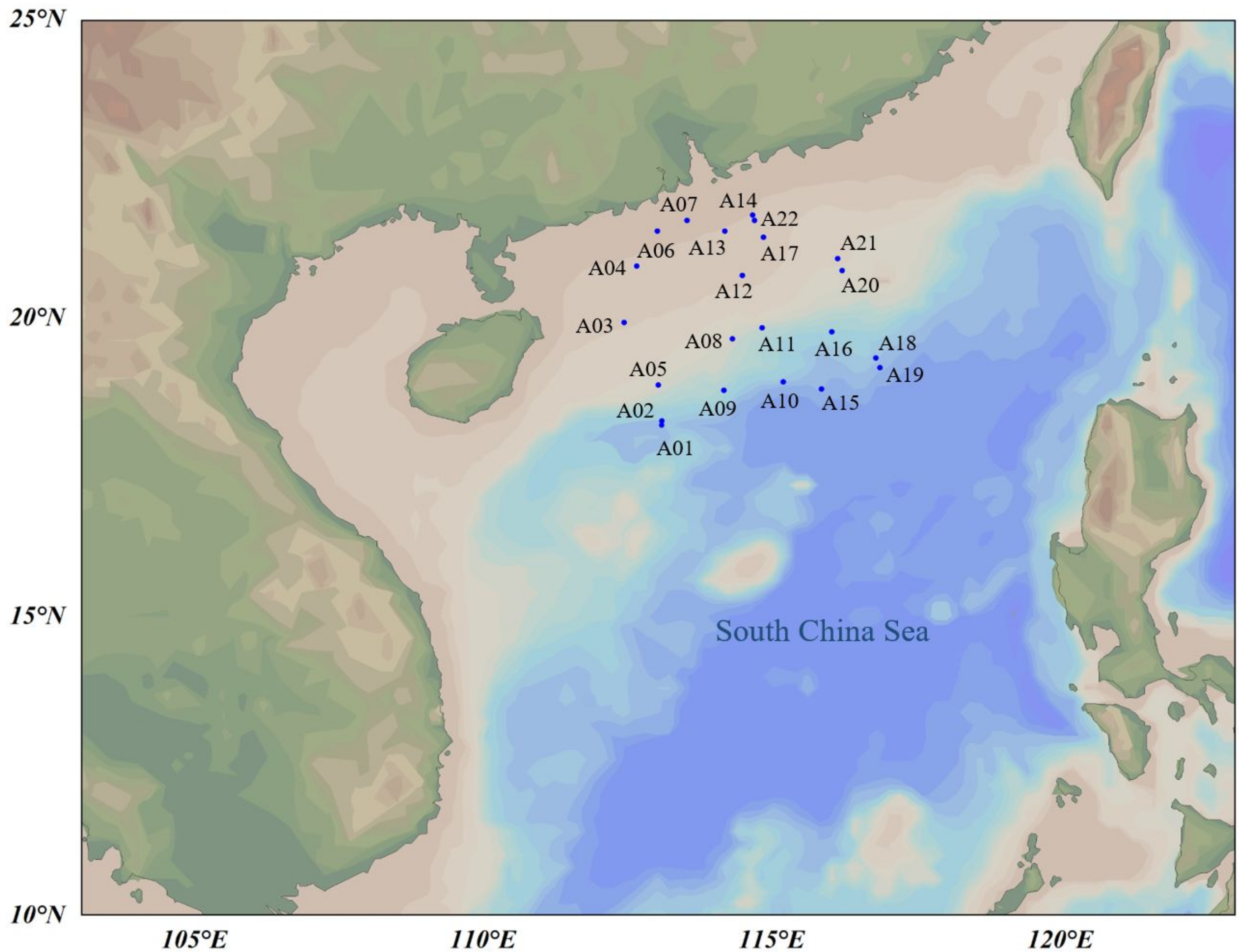


Figure S2. (A) Seawater sampling stations in the South China Sea, with the black dots marked at the starting position of each water sample. The color map shows the salinity profile in surface seawater; (B) general water mass circulation in the research area in summer

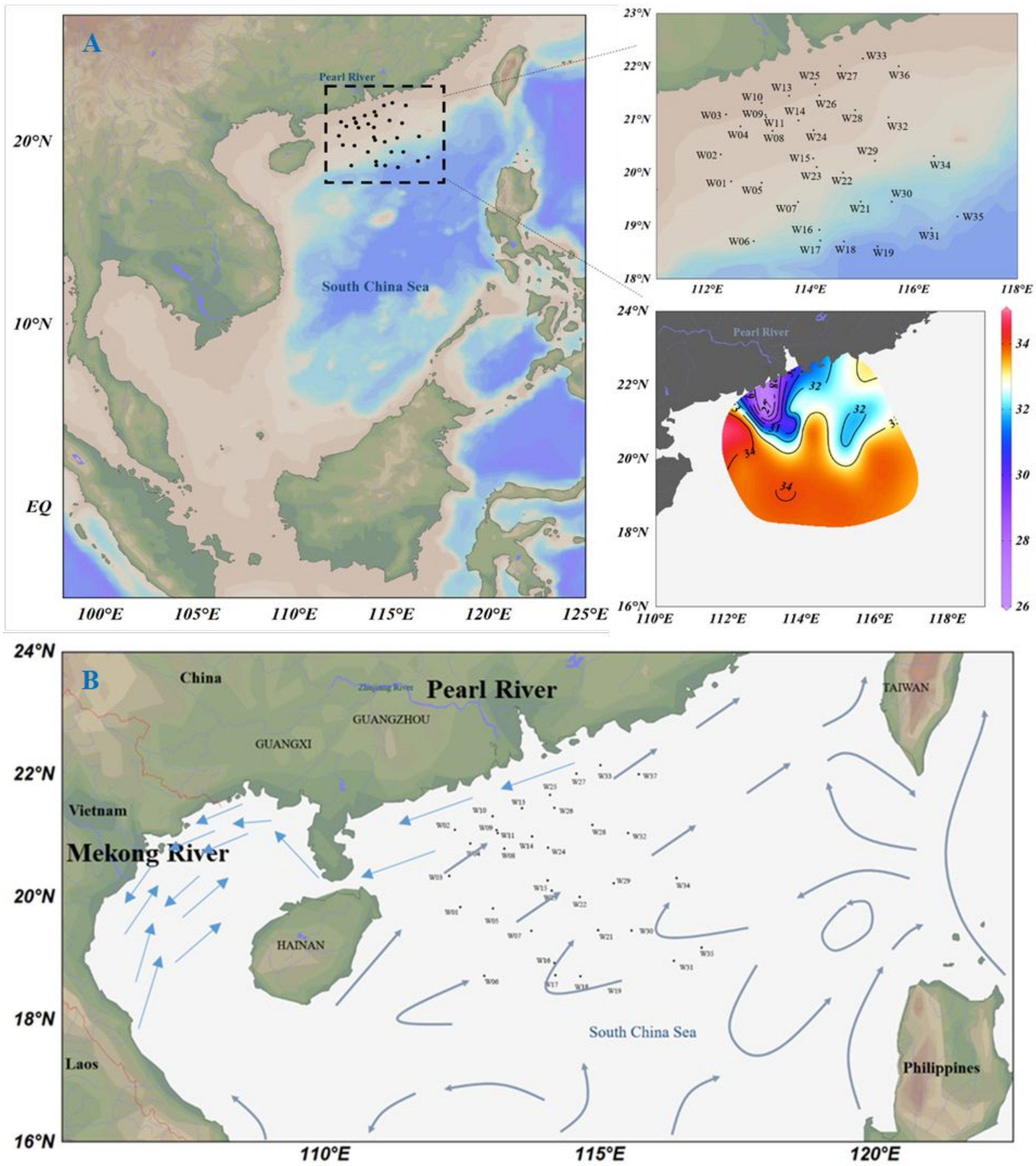


Figure S3A. High-volume air sampler is equipped with quartz fiber filter for atmospheric particles and a PUF/XAD-2 column for gaseous phase. The air sampler is placed at the upper deck and operated with headwind. The Filter and PUF-XAD-2 column are changed at sampling site. The PUF/XAD-2 column can be sealed with glass caps immediately after disconnected from the sampler to eliminate exposure time to the indoor air. The Quartz fiber filter is wrapped in a clean aluminum. PUF/XAD2 columns and filters are stored at -20°C;

S3B: The set-up of high-volume water sampler equipped with glass fiber filter and a XAD-2 column. The high-volume water sampler is connected to the ship-intake system with metal tubing. A glass fiber filter is closed in the filter holder to collect suspended particular matters and a XAD-2 column to retain organic chemicals in the dissolved phase. The seawater flows over the entire system at a flow rate of 0.2-1 L/min. The XAD-2 column is sealed with clips immediately after the sampling, and stored at 5°C. Glass fiber filter is wrapped in a clean aluminum foil and stored at -20°C

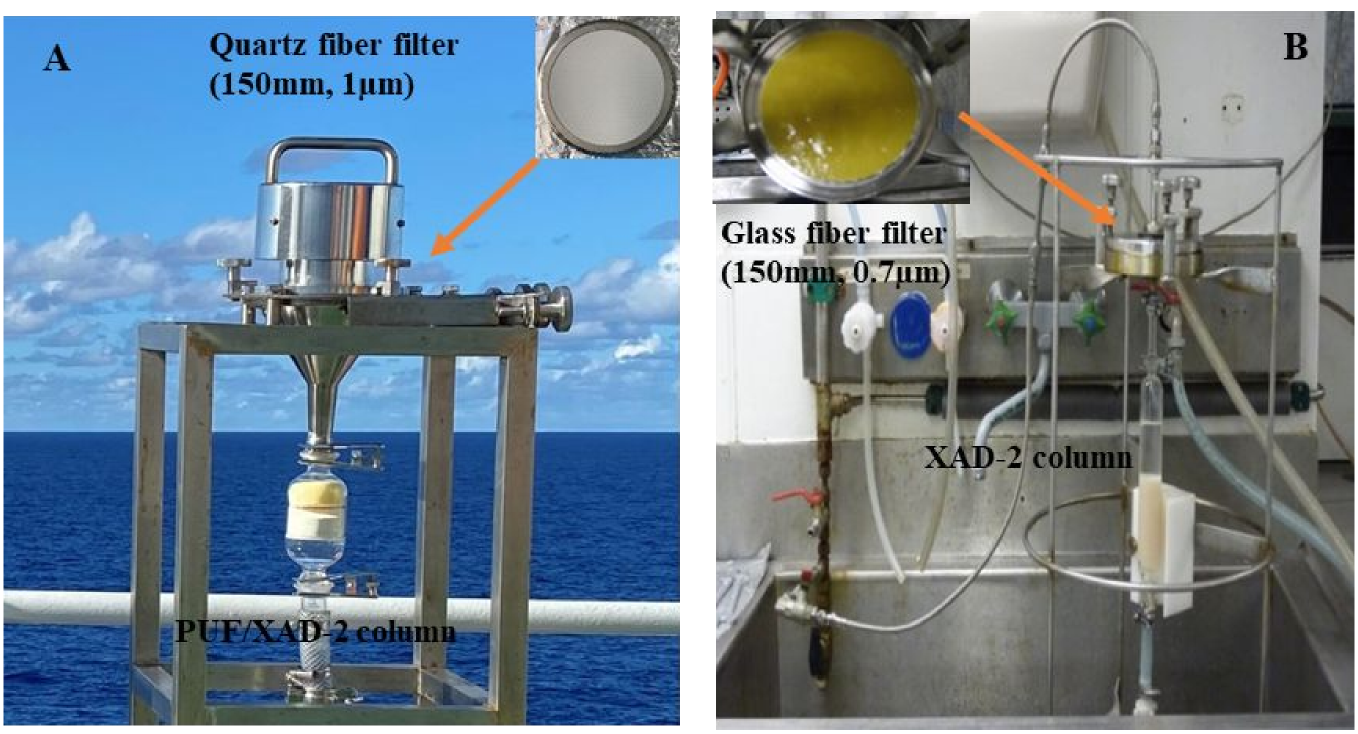
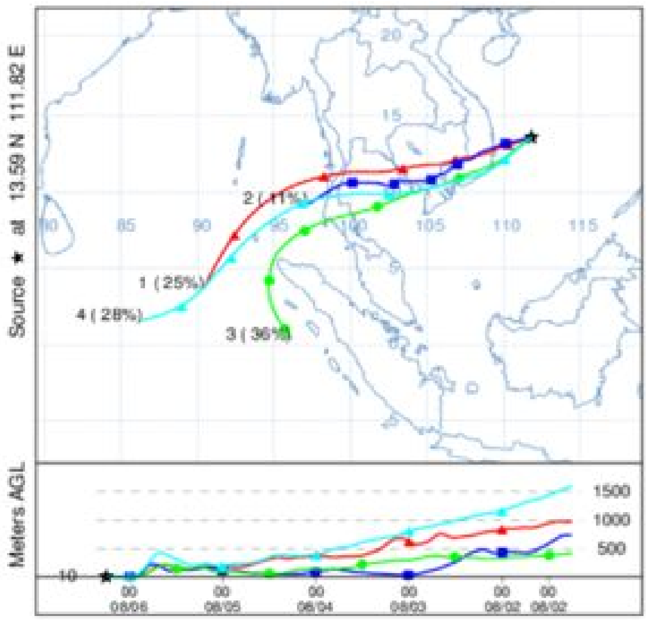


Figure S4. Air mass back trajectories of air samples. Colors indicate the origin of the air mass parcels



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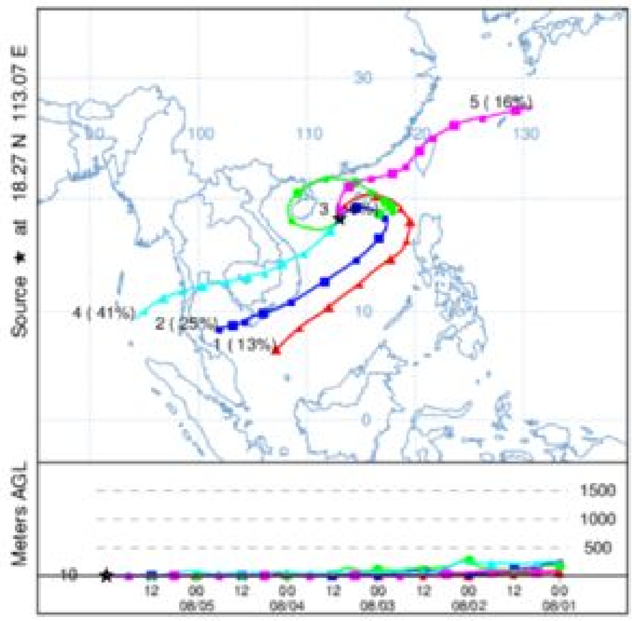

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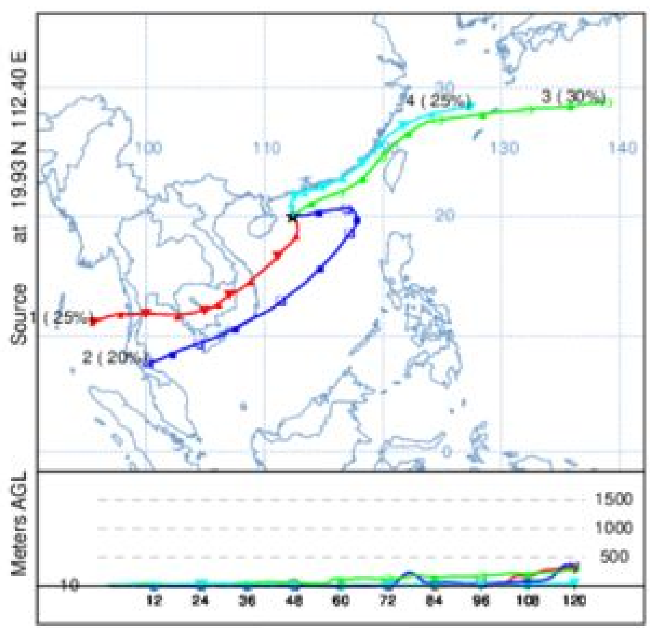

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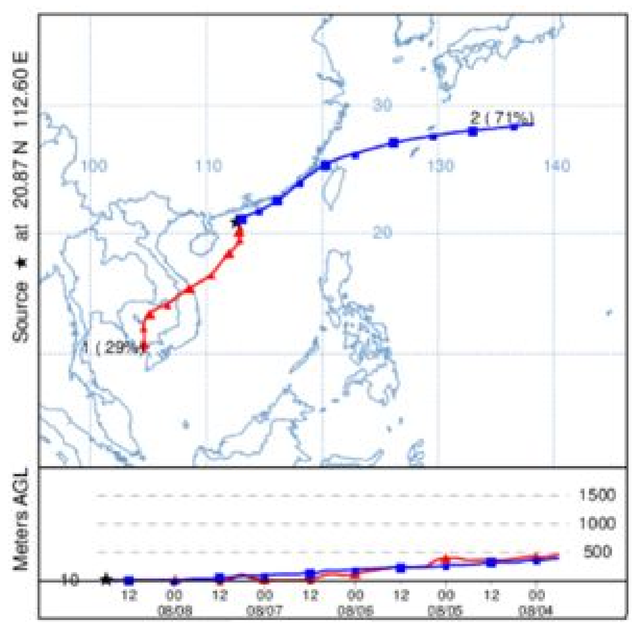

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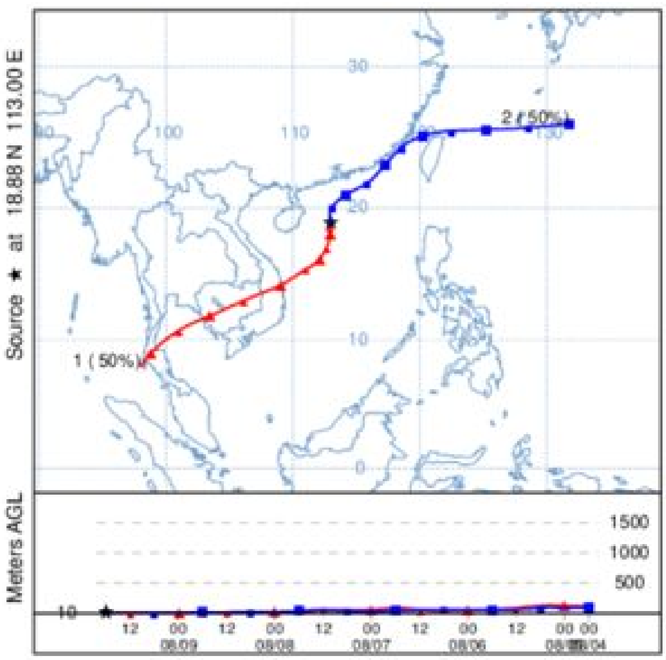

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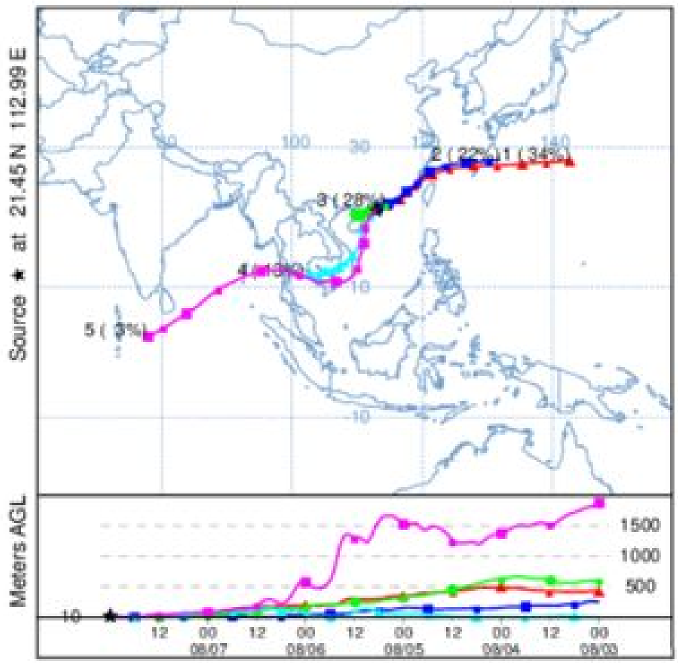

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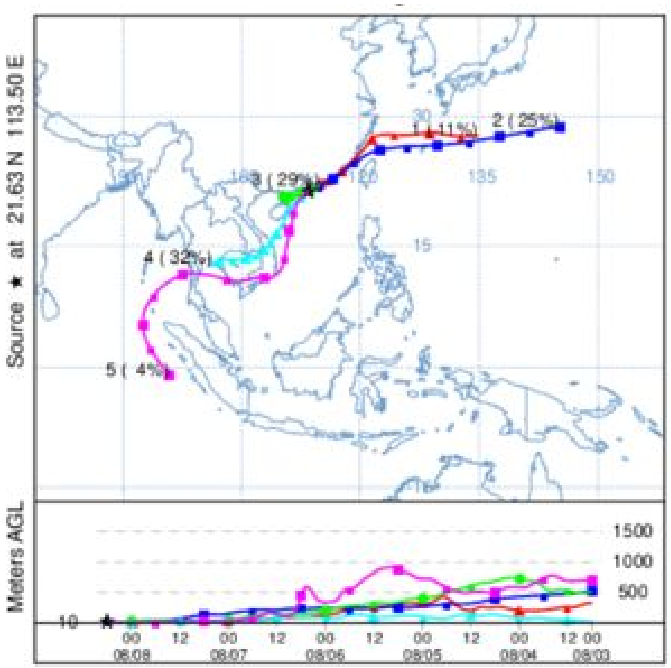

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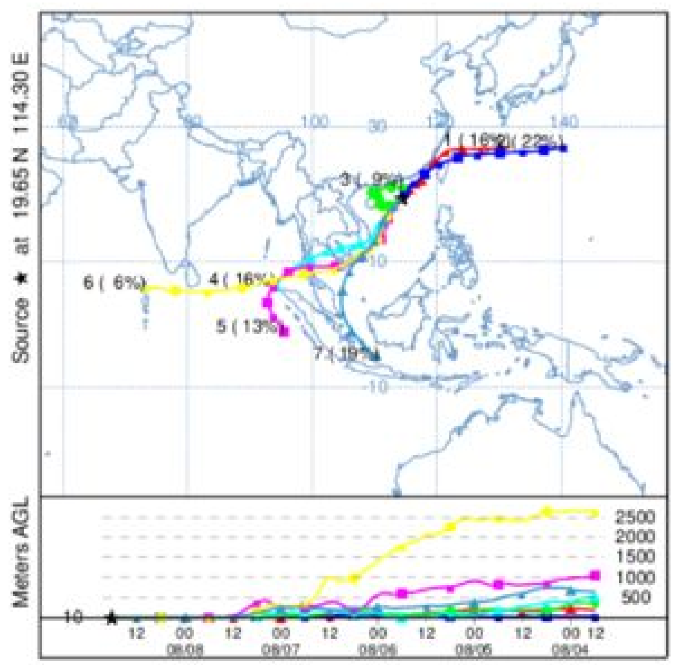

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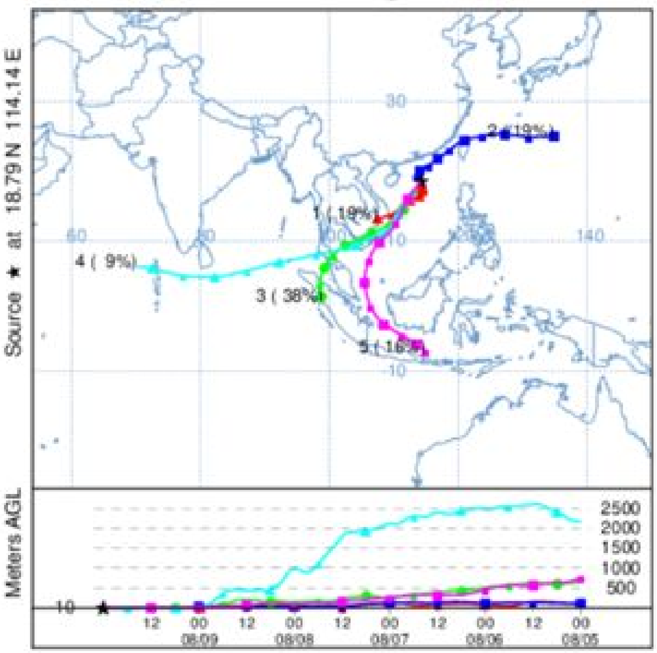

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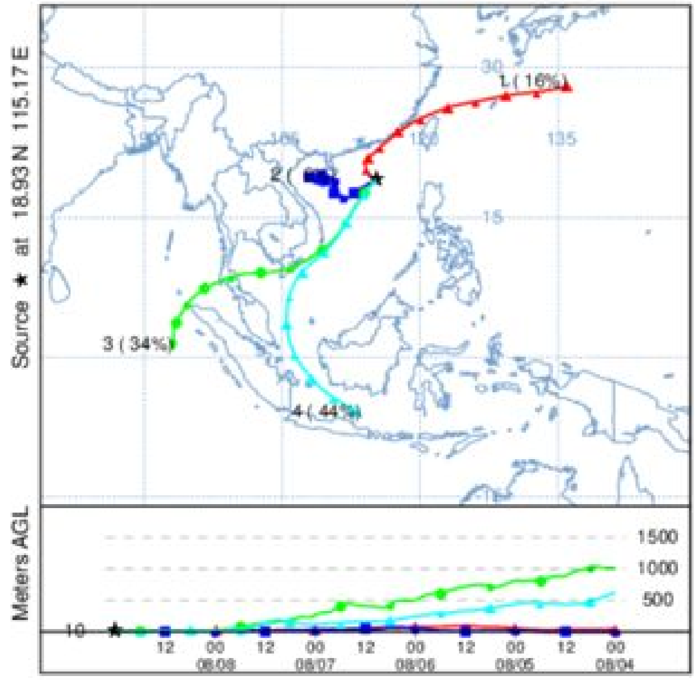

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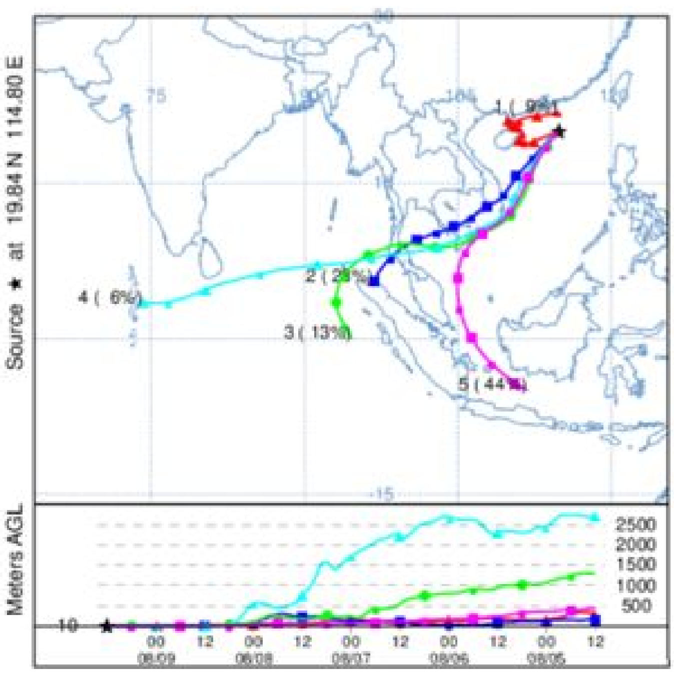

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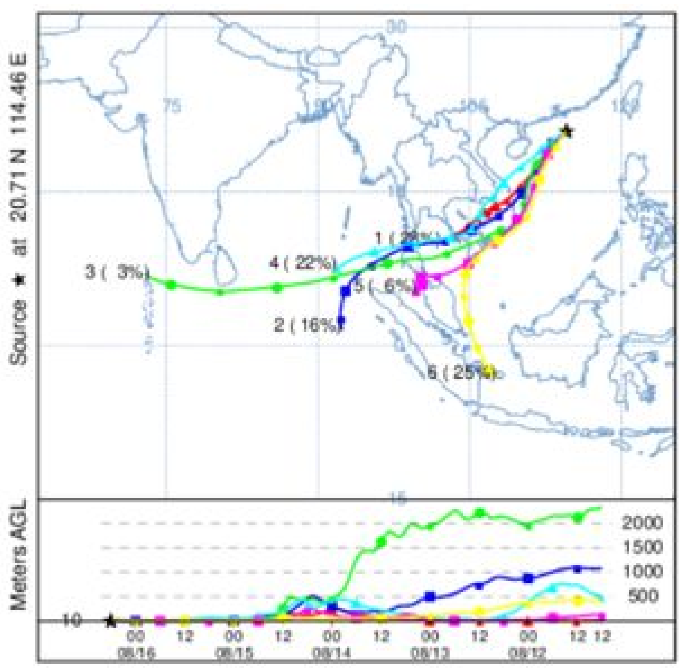

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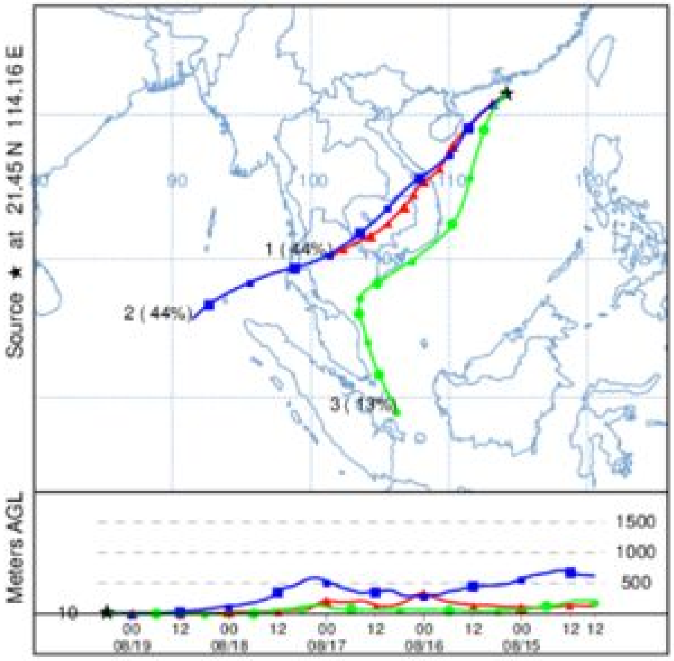

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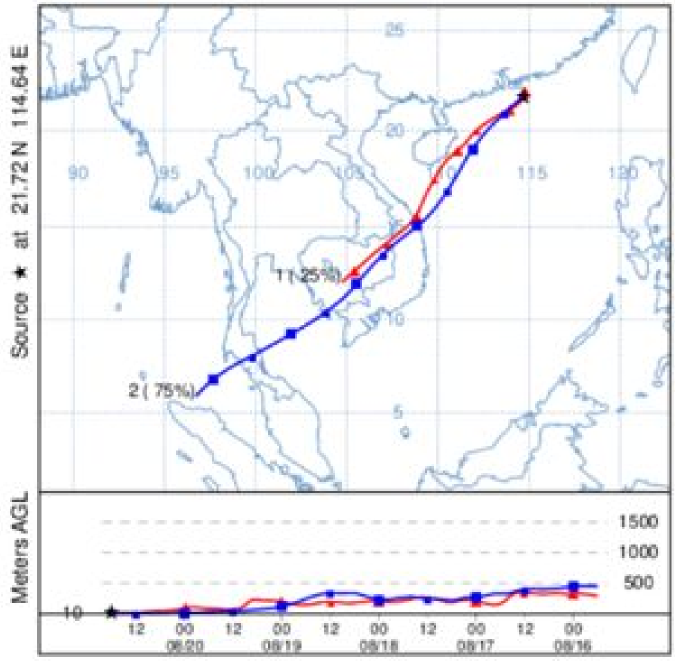

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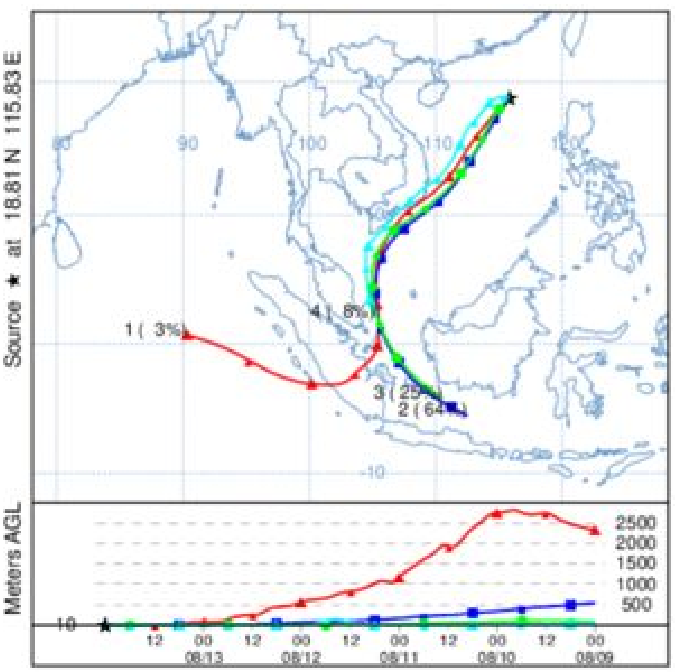

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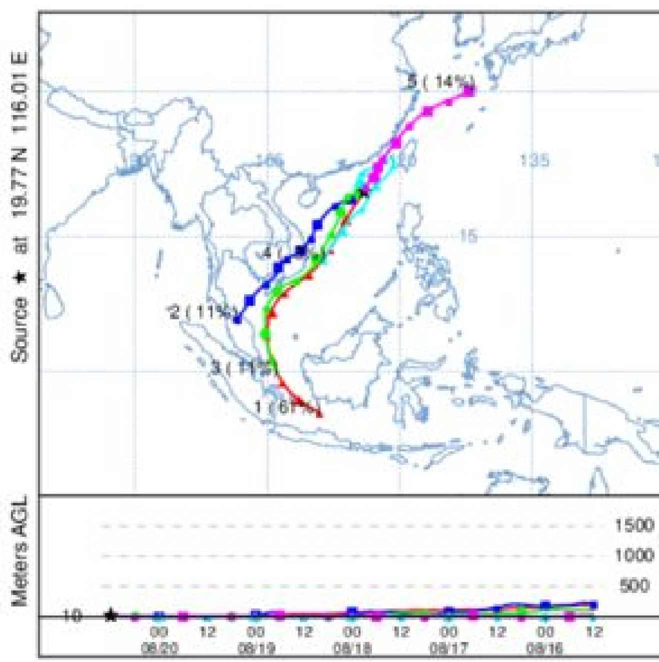

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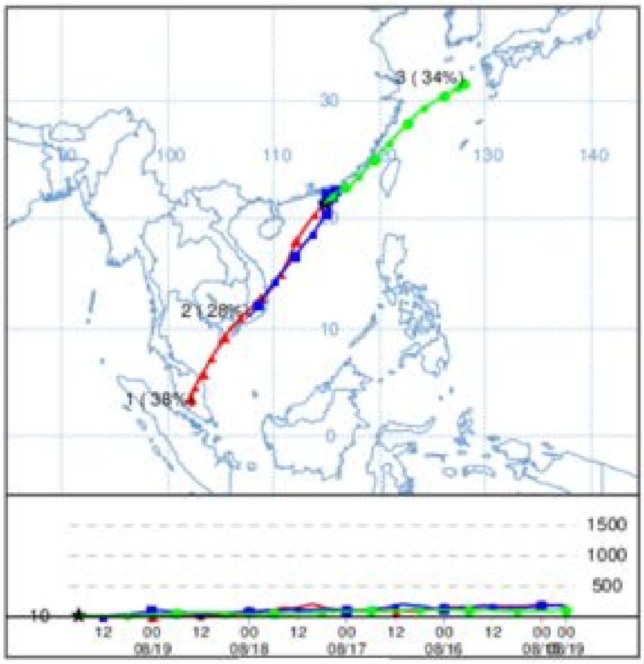

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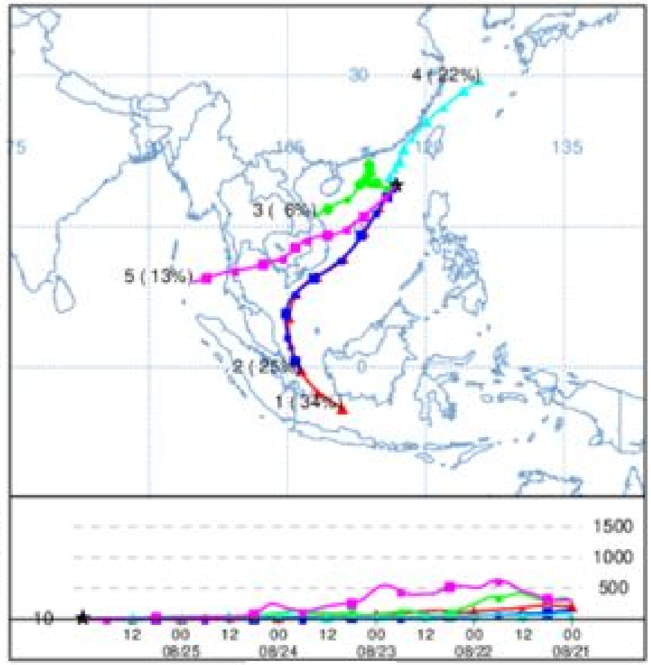

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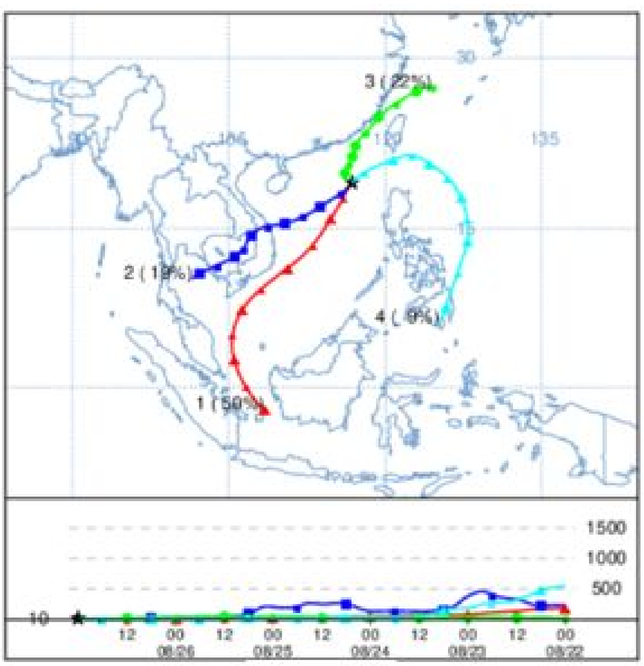

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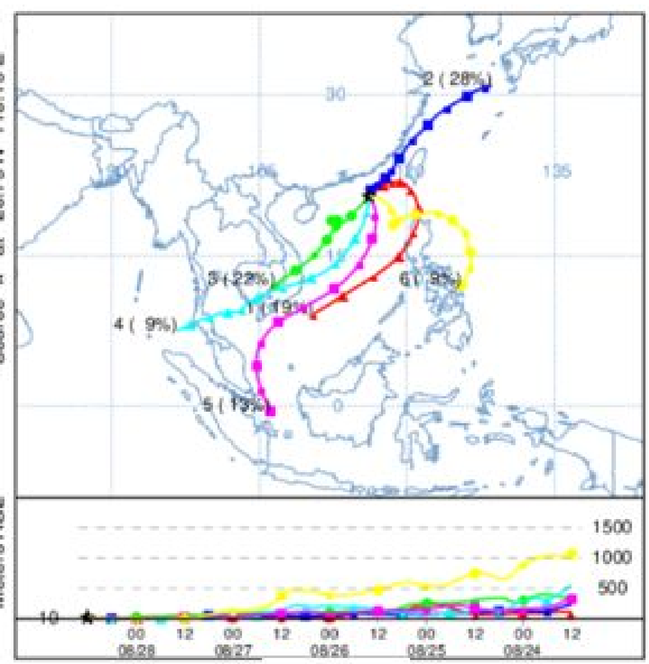

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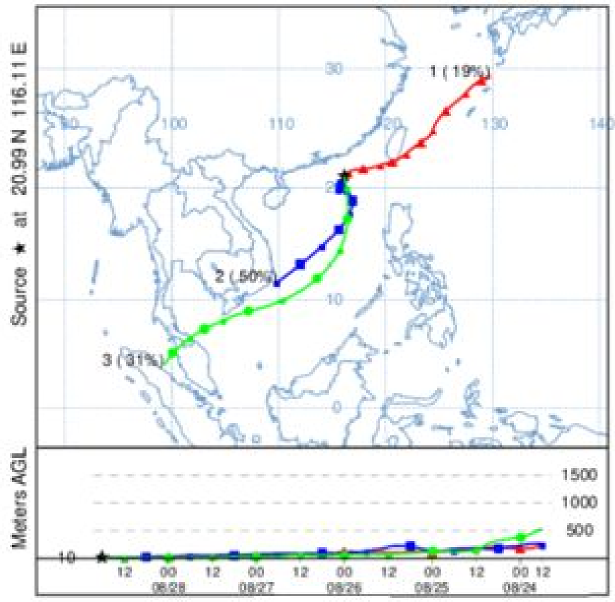
Source ★ at 20.99 N 116.11 E
1 (19%)
2 (50%)
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Meters AGL
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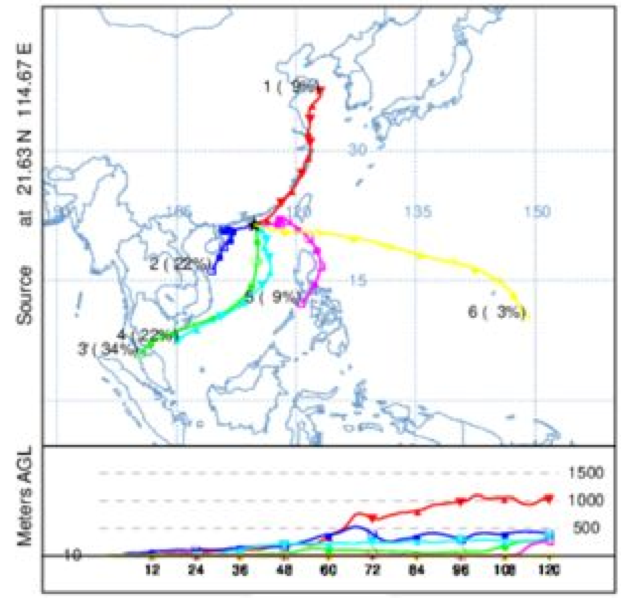
Source at 21.63 N 114.67 E
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5 (9%)
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Meters AGL
1500
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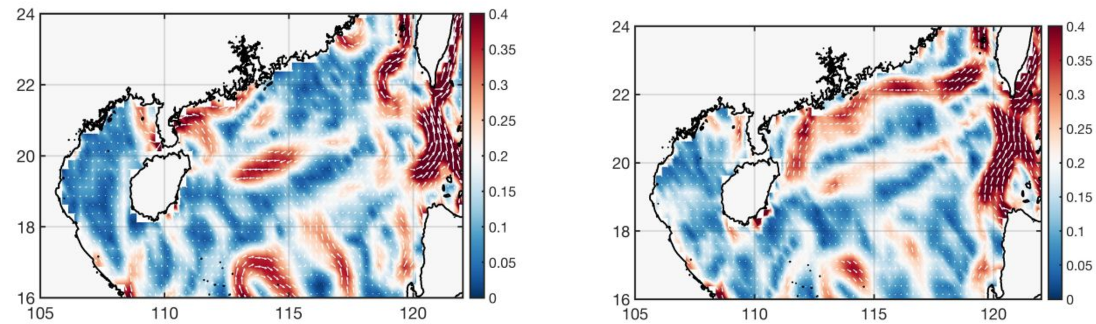
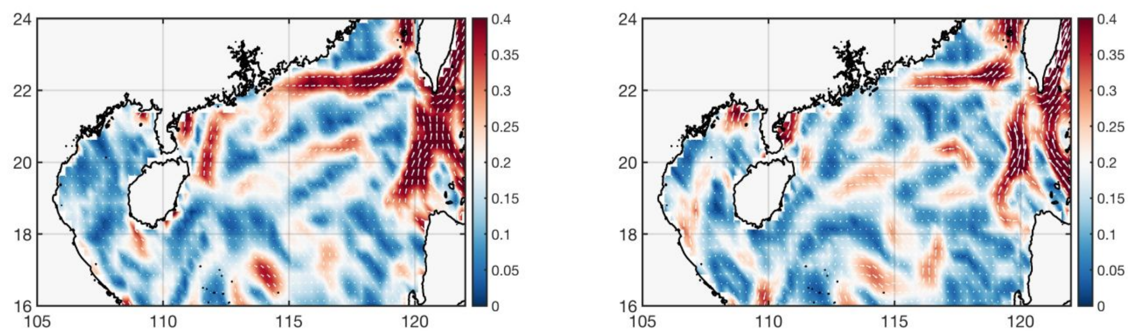
A22

Figure S5. The schematic diagrams of the flow field of the South China Sea in 7th (A) 13th (B), 19th (C) and 25th (D) of August 2019



B



D

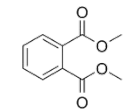
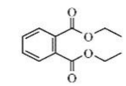
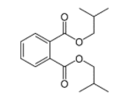
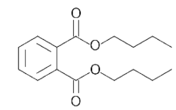
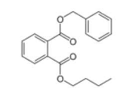
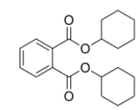
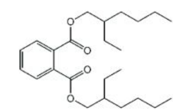
Table S 1. Detail information for air sampling

Air sample	Latitude (North, degree)	Longitude (East, degree)	Sampling Date	Time	Volume (m^3)	Air temperature (℃)	Humidity (%)	Wind speed (m/s)	Wind direction (degree)
A01	13.589	111.820	5-Aug-2019	0:37	382	29.1	73.8	11.3	244.5
A02	18.278	113.070	6-Aug-2019	7:30	373	29	82.4	9.9	336.8
A03	19.927	112.414	7-Aug-2019	11:08	293	28.8	83.7	7.2	50.2
A04	20.867	112.632	8-Aug-2019	10:37	269	29.2	80.8	4.8	232.2
A05	18.875	112.997	9-Aug-2019	11:28	326	29.5	82.7	6.3	205
A06	21.453	112.986	11-Aug-2019	11:03	341	29.6	83.4	8.2	219.5
A07	21.631	113.505	12-Aug-2019	11:03	348	29.6	84.5	8.2	221.6
A08	19.651	114.290	13-Aug-2019	10:12	344	29.6	84.2	8.4	219.1
A09	18.787	114.142	14-Aug-2019	10:54	280	29.7	75.3	8.9	229.6
A10	18.926	115.167	15-Aug-2019	10:54	316	29.2	82.8	11.1	201.3
A11	19.839	114.797	16-Aug-2019	10:54	278	29.6	83	7.5	198.8
A12	20.708	114.456	17-Aug-2019	11:05	645	29.4	83.8	7.6	214.4
A13	21.452	114.160	19-Aug-2019	10:39	290	29.4	74.8	6	223
A14	21.720	114.644	20-Aug-2019	11:00	535	28.6	74.4	8.3	94.3
A15	18.813	115.830	22-Aug-2019	10:39	339	26.6	87	9.7	314.9
A16	19.766	116.008	23-Aug-2019	11:46	356	28.9	80.3	3.4	26
A17	21.351	114.831	24-Aug-2019	11:46	504	30.3	79.9	12.5	242.3
A18	19.332	116.770	26-Aug-2019	11:00	310	29.2	81.7	3.9	141.6
A19	19.166	116.840	27-Aug-2019	11:00	531	28.7	81.7	7.2	35.3
A20	20.791	116.190	29-Aug-2019	11:00	345	28.5	81.3	7.8	124.7
A21	20.993	116.111	30-Aug-2019	11:06	313	28.5	78.1	4.6	108.8
A22	21.630	114.665	31-Aug-2019	11:15	300	28.7	79.4	7.9	81.5

Table S 2. Detail information for seawater sampling

Seawater Sample	Latitude (North, degree)	Longitude (East, degree)	Sampling Date	Volume (L)	Ta (℃)	Tw (℃)	Conductivity (S/m)	Depth (m)
W01	19.83	112.45	7-Aug-2019	100	28.8	29.9	5.64	113.2
W02	20.34	112.25	7-Aug-2019	135	28.5	29.6	5.65	83.4
W03	21.09	112.35	8-Aug-2019	171	29	29.4	5.64	48.8
W04	20.87	112.63	8-Aug-2019	178	29.4	29.6	5.67	60
W05	19.81	113.04	9-Aug-2019	193	29.4	30	5.67	154
W06	18.71	112.89	9-Aug-2019	177	29.4	30	5.67	900
W07	19.45	113.75	10-Aug-2019	244	29.8	29.8	5.67	472.5
W08	20.78	113.25	10-Aug-2019	196	29.3	30	5.4	81.1
W09	21.09	113.12	11-Aug-2019	36	29.8	30.3	4.74	60
W10	21.31	113.04	11-Aug-2019	169	29.8	29.8	4.57	42
W11	21.04	113.13	12-Aug-2019	84	30.3	30.4	4.98	44.3
W12	21.04	113.13	12-Aug-2019	119	30.3	30.4	4.98	44.3
W13	21.44	113.57	12-Aug-2019	68	29.7	29.6	5.18	42.2
W14	20.98	113.75	12-Aug-2019	105	29.7	30.4	4.83	85.6
W15	20.27	114.04	13-Aug-2019	310	29.8	29.6	5.58	121.4
W16	18.92	114.16	13-Aug-2019	199	29.5	29.8	5.66	1348.3
W17	18.72	114.18	14-Aug-2019	198	29.5	29.8	5.66	1859.1
W18	18.70	114.64	14-Aug-2019	126	29.2	29.7	5.65	3510
W19	18.61	115.29	15-Aug-2019	247	29.2	29.4	5.62	3694.5
W21	19.45	114.96	16-Aug-2019	195	29.9	29.7	5.62	1469.2
W22	20.00	114.62	16-Aug-2019	195	27.7	29.7	5.58	1469.2
W23	20.10	114.11	17-Aug-2019	165	29.6	29.8	5.61	187.5
W24	20.80	114.05	18-Aug-2019	165	29.6	30	5.68	89
W25	21.66	114.08	18-Aug-2019	217	27.7	29.6	5.45	51
W26	21.45	114.16	19-Aug-2019	210	29.2	29.9	5.42	55
W27	22.01	114.56	20-Aug-2019	143	28.4	29.2	5.3	40
W28	21.17	114.85	20-Aug-2019	158	28.4	29.7	5.46	95
W29	20.22	115.24	21-Aug-2019	185	26.7	29.4	5.35	468
W30	19.45	115.56	22-Aug-2019	258	28.7	29.3	5.56	2490
W31	18.95	116.33	23-Aug-2019	240	28.8	29.4	5.61	2732
W32	21.04	115.5	24-Aug-2019	220	29.4	29.8	5.34	125
W33	22.14	115	25-Aug-2019	250	27.5	29.9	5.44	51.8
W34	20.31	116.38	26-Aug-2019	138	29.2	29.4	5.58	961
W35	19.17	116.84	26-Aug-2019	180	29.2	29.5	5.6	3339
W36	19.17	116.84	27-Aug-2019	35	29.5	29.6	5.6	3340
W37	22.00	115.7	31-Aug-2019	100	29.2	29.9	5.54	80

Table S 3. Physiochemical properties of seven phthalate esters[9]

Chemical Name	Abbreviation	CAS	MW	Chemical formula	V_p (Pa)	S (mg/L)	H (pa.m^3/mol)	logK_{ow}	logK_{oa}	logKaw	Half-life time in air (d)	Chemical structure
Dimethyl phthalate	DMP	131-11-3	194.19	$C_{10}H_{10}O_4$	0.263	5220	$9.78x10^{-3}$	1.61	7.01	-5.4	14.4	
Diethyl phthalate	DEP	84-66-2	222.24	$C_{12}H_{14}O_4$	$6.48x10^{-2}$	591	$2.44x10^{-2}$	2.54	7.55	-5.01	2.39	
Diisobutyl phthalate	DiBP	84-69-5	278.35	$C_{16}H_{22}O_4$	$4.73x10^{-3}$	9.9	0.13	4.27	8.54	-4.27	0.58	
Dibutyl phthalate	DnBP	84-74-2	278.34	$C_{16}H_{22}O_4$	$4.73x10^{-3}$	9.9	0.13	4.27	8.54	-4.27	0.89	
Benzylbutyl phthalate	BBP	85-68-7	312.37	$C_{19}H_{20}O_4$	$2.49x10^{-3}$	3.8	0.205	4.70	8.78	-4.08	0.75	
Dicyclohexyl phthalate	DCHP	84-61-7	330.42	$C_{20}H_{26}O_4$	$6.1x10^{-4}$	$4.1x10^{-2}$	4.92	6.2	11.59	-5.39	0.22	
Bis(2-ethylhexyl) phthalate	DEHP	117-81-7	390.56	$C_{24}H_{38}O_4$	$2.52x10^{-5}$	$2.49x10^{-3}$	3.95	7.73	10.53	-2.80	0.38	

V_p: vapor pressure at 25°C; S: water solubility at 25°C; H: Henry's law constants at 25°C; logKow: log Octanol-Air coefficient at 25°C; logK_{oa}: log Octanol-Air coefficient at 25°C; logKaw: log air-water transfer coefficient; Half-life time in air (d) (exclude DiBP and DCHP)[10]; Half-live time in air for DiBP and DCHP: (Calculated by EPIWEB 4.1). Physiochemical properties of DCHP was Calculated with EPIWEB 4.1.

Table S 4. Information about the chemicals and material applied in this work

Chemical and material	**Description**	**Supplier**
Dichloromethane	Pico grade	Promochem, Wesel, Germany
n-Hexane	Pico grade	Promochem, Wesel, Germany
Methanol	Pico grade	Promochem, Wesel, Germany
Acetone	Pico grade	Promochem, Wesel, Germany
Dimethyl phthalate-3,4,5,6-d4	Ring-d4, 98%, 100µg/mL	LGC, Wesel, Germany
Diethyl phthalate-3,4,5,6-d4	Ring-d4, 98%, 100µg/mL	LGC, Wesel, Germany
Dibutyl phthalate-3,4,5,6-d4	Ring-d4, 98%, 100µg/mL	LGC, Wesel, Germany
Bis(2-ethylhexyl)phthalate-3,4,5,6-d4	Ring-d4, 98%, 100µg/mL	LGC, Wesel, Germany
2, 2',3,3',4,5,5',6,6'-Nonachlorobiphenyl (13C12)	99%, 2µg/mL	Cambridge Isotope Laboratories, UK
Phthalate esters Analytes Mix 3 Content: DMP, DEP, DiBP, DnBP, BBP, DCHP, DEHP and other 10 PAEs	1000µg/mL	LGC, Wesel, Germany
Silica gel	0.063-0.2mm	Macherey Nagel, Düren, Germany
Anhydrous sodium sulphate	99%	Merck, Darmstadt, Germany
Amberlite XAD-2	20-60 mesh	Merck, Darmstadt, Germany

Table S 5. Agilent 7890 GC-MS/MS conditions

Parameter	Value
Analytical column	2 set Agilent HP5-ms, 250 µm × 15 m, 0.25 µm
Sample injection mode	1 µL Pulsed splitless using Multimode Inlet (MMI)
Injection port liner	Agilent 200 µL dimpled, single-taper liner
Injection temperature program	20 °C (0.2 minutes), 300 °C/min to 300 ℃
Injection pulsed pressure	25 psi (1.9 minutes)
Purge flow to spit vent	50 mL/min (2.0 minutes)
Carrier gas	Helium
Carrier gas flow	1.2 mL/min
Oven temperature program	50 °C for 2 min, 20 °C/min to 80 °C, 5 °C/min to 250 °C, 15 °C/min to 300 °C (14 min)
Post run	310 °C for 3 min
Backflush	3.0 ml/min
Mass spectrometer	Agilent 7000C with electron impact ionization source operated in multiple reaction monitoring mode (MRM)
Ionization mode	Positive
Source temperature	280 °C
Quadrupole 1 and 2 temperature	150 °C
Transfer line temperature	280 °C

Table S 6. Detail information for the determination of PAEs using GC-MS/MS

PAE	Quantifier ions (m/z)	Qualifier ions (m/z)	Collision energy (eV)	Retention time (min)	Instrumental detection limit(pg)
DMP	163>77	163>135	15	16.963	0.02
DEP	149>93	149>121	15	20.258	0.03
DiBP	149>93	149>121	15	26.105	0.04
DnBP	149>93	149>121	15	27.956	0.04
BBP	149>93	149>121	15	34.854	0.1
DCHP	149>93	149>121	15	37.532	0.1
DEHP	149>93	149>121	15	37.955	0.2
d4-DMP	167>81	167>139	15	16.933	-
d4-DEP	153>97	153>125	15	20.232	-
d4-DnBP	153>97	153>125	15	27.937	-
d4-DEHP	153>97	153>125	15	37.943	-

Table S 7. The field blanks and the method detection limits of PAEs in PUF/XAD-2, XAD-2 columns, QFF and GFF filters

Sample material	**PUF/XAD-2 column (gas)**		**QFF (particle)**		**XAD-2 column (dissolved phase)**		**GFF (particular matter)**	
PAE	**Blank (mean±SD) (ng/m^3)**	**MDL (ng/m^3)**	**Blank (mean±SD) (ng/m^3)**	**MDL (ng/m^3)**	**Blank (mean±SD) (ng/L)**	**MDL (ng/L)**	**Blank (mean±SD) (ng/L)**	**MDL (ng/L)**
DMP	0.017±0.006	0.036	0.002±0.001	0.004	0.002±0.001	0.004	0.005±0.002	0.011
DEP	0.014±0.002	0.022	0.008±0.004	0.021	0.008±0.004	0.021	0.021±0.008	0.045
DiBP	0.020±0.007	0.042	0.005±0.001	0.007	0.005±0.001	0.007	0.045±0.011	0.077
DnBP	0.037±0.011	0.068	0.006±0.002	0.011	0.006±0.002	0.011	0.015±0.001	0.017
BBP	0.060±0.010	0.090	0.001±0.000	0.001	0.001±0.000	0.001	0.001±0.001	0.002
DCHP	0.001±0.000	0.002	0.001±0.000	0.001	0.001±0.000	0.001	0.001±0.000	0.001
DEHP	0.14±0.002	0.15	0.074±0.031	0.166	0.074±0.031	0.17	0.17±0.099	0.47

Table S 8. Concentrations of PAEs in the atmosphere over the South China Sea. The MDLs of PAEs calculated with individual sampling volumes are given in brackets for the calculated concentrations less than MDLs.

Air sample	DMP	DEP	DiBP	DnBP	BBP (MDL)	DCHP (MDL)	DEHP	$\sum_7$PAEs
Gaseous phase (Cg, ng/m³)								
A01	0.45	0.10	1.37	0.94	0.02 (0.09)	0.00 (0.002)	2.80	5.68
A02	0.26	0.25	0.74	0.34	0.06 (0.09)	0.00 (0.002)	1.42	3.07
A03	1.60	0.14	2.93	2.13	0.09 (0.11)	0.002	0.47	7.36
A04	0.89	0.17	3.46	2.48	0.03 (0.12)	0.024	0.86	7.91
A05	0.45	0.26	1.46	1.13	0.03 (0.10)	0.005	0.68	4.00
A06	0.27	0.13	0.58	0.57	0.07 (0.10)	0.001 (0.002)	0.67	2.28
A07	0.32	0.17	1.93	1.63	0.03 (0.09)	0.003	0.70	4.78
A08	0.15	0.06	0.29	0.35	0.05 (0.10)	0.001 (0.002)	0.46	1.37
A09	0.35	0.13	1.15	0.82	0.02 (0.12)	0.013	0.63	3.12
A10	0.19	0.06	0.15	0.09	0.03 (0.10)	0.001 (0.002)	0.25	0.76
A11	0.29	0.26	1.76	1.19	0.01 (0.12)	0.001 (0.003)	0.50	4.00
A12	0.28	0.14	4.02	2.90	0.08	0.006	0.36	7.79
A13	0.68	0.15	2.31	1.81	0.08 (0.11)	0.002 (0.003)	0.85	5.88
A14	0.65	0.15	3.58	3.00	0.03 (0.06)	0.004	0.79	8.21
A15	0.36	0.05	0.46	0.38	0.02 (0.10)	0.002	0.31	1.58
A16	1.08	0.12	0.05	0.15	0.04 (0.09)	0.001 (0.002)	0.22	1.68
A17	1.53	0.39	1.50	1.96	0.02 (0.06)	0.002	1.08	6.48
A18	0.12	0.06	1.10	0.78	0.07 (0.10)	0.001 (0.002)	0.19	2.32
A19	0.14	0.06	1.30	1.12	0.04 (0.06)	0.003	0.34	3.00
A20	0.24	0.08	0.16	0.12	0.01 (0.10)	0.00 (0.002)	0.22	0.83
A21	0.47	0.19	1.26	1.06	0.05 (0.10)	0.002	0.30	3.33
A22	0.25	0.20	4.63	4.26	0.07 (0.10)	0.012	0.55	9.98
Mean	0.50	0.15	1.64	1.33	0.04 (0.09)	0.00 (0.002)	0.67	4.34
S.D.	0.42	0.08	1.33	1.10	0.03	0.01	0.57	2.70
Median	0.34	0.14	1.33	1.09	0.03 (0.09)	0.00 (0.002)	0.52	3.67
Max	1.60	0.39	4.63	4.26	0.09 (0.09)	0.02	2.80	9.98
Min	0.12	0.05	0.05	0.09	0.01 (0.09)	0.00 (0.002)	0.19	0.76
Particle phase (Cp, ng/m³)								
A01	0.01	0.02	0.01	0.02	0.0002 (0.001)	0.000 (0.001)	0.54	0.60
A02	0.01	0.01	0.10	0.06	0.001	0.000 (0.001)	1.15	1.32
A03	0.09	0.21	8.08	5.31	0.001	0.0013	1.04	14.72
A04	0.03	0.07	6.35	3.60	0.002	0.0102	0.49	10.56
A05	0.02	0.08	5.08	2.74	0.000 (0.001)	0.0011	0.60	8.52
A06	0.02	0.04	2.94	3.41	0.003	0.000 (0.001)	0.39	6.81
A07	0.01	0.04	0.36	1.65	0.005	0.0079	0.40	2.47
A08	0.01	0.04	0.96	1.24	0.002	0.0025	0.31	2.56
A09	0.02	0.07	2.87	2.63	0.002	0.0139	0.39	6.00
A10	0.02	0.05	7.97	6.57	0.005	0.0015	0.73	15.34
A11	0.01	0.05	3.05	3.54	0.003	0.0043	0.53	7.20
A12	0.01	0.02	0.31	0.33	0.002	0.0012	0.23	0.90
A13	0.03	0.03	0.36	0.48	0.001	0.0024	0.44	1.34
A14	0.02	0.07	0.10	0.24	0.004	0.0051	0.50	0.93
A15	0.02	0.14	3.20	2.38	0.008	0.0062	0.47	6.22
A16	0.16	0.14	0.13	0.17	0.003	0.001 (0.001)	0.55	1.16
A17	0.01	0.06	0.15	0.23	0.003	0.0010	0.60	1.06
A18	0.03	0.13	0.30	0.31	0.001	0.0005	0.68	1.46
A19	0.01	0.02	0.01	0.03	0.000 (0.001)	0.0008	0.15	0.21
A20	0.03	0.07	3.67	2.49	0.002	0.001 (0.001)	0.32	6.58
A21	0.04	0.20	10.50	9.44	0.005	0.020	0.80	21.00
A22	0.01	0.03	0.04	0.06	0.000 (0.001)	0.001 (0.001)	0.34	0.48
Mean	0.03	0.07	2.57	2.13	0.002	0.004	0.53	5.34
S.D.	0.03	0.06	3.17	2.47	0.002	0.005	0.24	5.73
Median	0.02	0.06	0.66	1.44	0.002	0.001	0.50	2.51
Max	0.16	0.21	10.50	9.44	0.008	0.020	1.15	21.00
Min	0.00	0.01	0.01	0.02	0.000 (0.001)	0.000 (0.001)	0.15	0.21
Sum of Cg+Cp (ng/m³)								
A01	0.46	0.12	1.38	0.96	0.02	0.00	3.34	6.28
A02	0.26	0.26	0.84	0.40	0.06	0.00	2.57	4.39
A03	1.69	0.35	11.01	7.44	0.09	0.00	1.51	22.08
A04	0.92	0.24	9.81	6.08	0.03	0.03	1.35	18.47
A05	0.47	0.34	6.54	3.87	0.03	0.00	1.28	12.52
A06	0.29	0.17	3.52	3.98	0.07	0.00	1.06	9.09
A07	0.33	0.21	2.29	3.28	0.03	0.01	1.10	7.25
A08	0.16	0.10	1.25	1.59	0.05	0.00	0.77	3.93

A09	0.37	0.20	4.02	3.45	0.02	0.02	1.02	9.12
A10	0.21	0.11	8.12	6.66	0.04	0.00	0.98	16.10
A11	0.30	0.31	4.81	4.73	0.01	0.00	1.03	11.20
A12	0.29	0.16	4.33	3.23	0.08	0.01	0.59	8.69
A13	0.71	0.18	2.67	2.29	0.08	0.00	1.29	7.22
A14	0.67	0.22	3.68	3.24	0.03	0.01	1.29	9.14
A15	0.38	0.19	3.66	2.76	0.03	0.01	0.78	7.80
A16	1.24	0.26	0.18	0.32	0.04	0.00	0.77	2.84
A17	1.54	0.45	1.65	2.19	0.02	0.00	1.68	7.54
A18	0.15	0.19	1.40	1.09	0.07	0.00	0.87	3.78
A19	0.15	0.08	1.31	1.15	0.04	0.00	0.49	3.21
A20	0.27	0.15	3.83	2.61	0.01	0.00	0.54	7.41
A21	0.51	0.39	11.76	10.50	0.05	0.02	1.10	24.33
A22	0.26	0.23	4.67	4.32	0.07	0.01	0.89	10.46
Mean	0.53	0.22	4.21	3.46	0.05	0.01	1.20	9.67
S.D.	0.44	0.10	3.31	2.48	0.02	0.01	0.66	5.86
Median	0.35	0.20	3.67	3.23	0.04	0.00	1.05	8.24
Max	1.69	0.45	11.76	10.50	0.09	0.03	3.34	24.33
Min	0.15	0.08	0.18	0.32	0.01	0.00	0.49	2.84

Table S 9. Particle-bound fractions (ø) of PAEs in air samples from the South China Sea. The particle-bound fractions are quite uncertain because BBP and DCHP were below MDLs in most particle samples.

Sample	DMP	DEP	DiBP	DnBP	BBP	DCHP	DEHP	PEs
A01	0.01	0.17	0.01	0.02	0.01	0.02	0.16	0.10
A02	0.02	0.05	0.11	0.16	0.01	0.49	0.45	0.30
A03	0.05	0.59	0.73	0.71	0.01	0.42	0.69	0.67
A04	0.03	0.29	0.65	0.59	0.06	0.30	0.36	0.57
A05	0.03	0.24	0.78	0.71	0.01	0.19	0.47	0.68
A06	0.07	0.23	0.84	0.86	0.04	0.29	0.37	0.75
A07	0.03	0.18	0.16	0.50	0.13	0.72	0.36	0.34
A08	0.06	0.40	0.77	0.78	0.03	0.65	0.40	0.65
A09	0.06	0.35	0.71	0.76	0.09	0.52	0.38	0.66
A10	0.08	0.42	0.98	0.99	0.15	0.72	0.75	0.95
A11	0.05	0.16	0.63	0.75	0.18	0.79	0.52	0.64
A12	0.03	0.10	0.07	0.10	0.02	0.17	0.40	0.10
A13	0.04	0.18	0.13	0.21	0.02	0.54	0.34	0.19
A14	0.03	0.32	0.03	0.07	0.13	0.54	0.39	0.10
A15	0.05	0.72	0.87	0.86	0.28	0.76	0.60	0.80
A16	0.13	0.54	0.70	0.52	0.06	0.36	0.72	0.41
A17	0.01	0.14	0.09	0.11	0.12	0.37	0.36	0.14
A18	0.2	0.68	0.22	0.29	0.02	0.42	0.78	0.39
A19	0.05	0.24	0.01	0.02	0.01	0.21	0.30	0.07
A20	0.1	0.46	0.96	0.95	0.13	0.86	0.59	0.89
A21	0.08	0.52	0.89	0.90	0.09	0.91	0.73	0.86
A22	0.05	0.13	0.01	0.01	0.01	0.06	0.38	0.05
Mean	0.06	0.32	0.47	0.49	0.07	0.47	0.48	0.47
S.D.	0.04	0.19	0.37	0.35	0.07	0.26	0.17	0.30

Table S 10. Washout ratio (W) calculated with particle bound fraction following eq. $W = (1-ø)(RT/H)+øW_p$. BBP and DCHP were excluded because of low detection frequency.

Washout ratio (W)	DMP	DEP	DiBP	DnBP	DEHP
A01	250001	88055	18650	18661	3784
A02	249049	97693	18794	18851	9284
A03	241086	53620	19638	19610	13992
A04	246568	77786	19520	19444	7669
A05	245343	81678	19696	19603	9745
A06	237432	83066	19777	19805	7783
A07	245799	86685	18854	19323	7621
A08	238486	69124	19680	19701	8411
A09	239887	73298	19610	19677	8006
A10	235434	67580	19975	19983	15097
A11	242910	88639	19502	19658	10631
A12	246794	93344	18734	18777	8327
A13	244920	86717	18820	18923	7234
A14	247213	75085	18674	18738	8111
A15	241118	42584	19830	19812	12229
A16	222975	57545	19594	19352	14479
A17	251596	90011	18759	18782	7559
A18	207786	46031	18934	19031	15690
A19	241474	82131	18647	18670	6495
A20	229263	64022	19945	19937	12013
A21	235419	59487	19854	19862	14700
A22	242384	91108	18649	18656	8038
Max	251596	97693	19975	19983	15690
Min	207786	42584	18647	18656	3784
Mean	240134	75241	19279	19312	9859
SD	9895	15795	510	480	3261
Median	241929	79732	19511	19398	8369

Table S 11. Concentrations of PAEs in the seawater from the South China Sea. The MDLs of PAEs calculated with individual sampling volumes are given in brackets for the calculated concentrations less than MDLs.

Sample	DMP	DEP	DiBP	DnBP	BBP	DCHP	DEHP	PAEs
PAE in dissolved phase (C_W, ng/L)								
W01	0.20	0.46	0.25	1.08	0.014	0.001 (0.002)	1.53	3.54
W02	0.15	0.27	0.21	1.38	0.008	0.001 (0.001)	1.39	3.40
W03	0.41	0.30	0.14	0.73	0.006	0.001 (0.001)	0.77	2.36
W04	0.12	0.14	0.18	0.49	0.005	0.001 (0.001)	0.93	1.85
W05	0.20	0.25	0.09	0.30	0.004	0.000 (0.001)	1.32	2.16
W06	0.24	0.14	0.11	0.50	0.003	0.001 (0.001)	0.63	1.64
W07	0.13	0.26	0.09	0.28	0.003	0.000 (0.001)	0.98	1.74
W08	1.60	0.93	0.31	0.21	0.006	0.013	0.48	3.55
W09	1.14	0.84	0.25	1.02	0.107	0.001 (0.005)	3.52	6.88
W10	1.30	0.57	0.13	0.17	0.027	0.007	2.03	4.23
W11	1.16	0.77	0.20	0.43	0.012	0.001 (0.002)	2.68	5.25
W12	0.77	0.35	0.08	0.20	0.003	0.000 (0.001)	1.01	2.42
W13	1.30	0.98	0.08	0.20	0.005	0.000 (0.002)	2.87	5.43
W14	0.25	0.43	0.09	0.22	0.008	0.000 (0.002)	3.30	4.30
W15	0.14	0.22	0.05	0.13	0.003	0.000 (0.001)	0.70	1.23
W16	0.14	0.19	0.07	0.17	0.020	0.001 (0.001)	1.04	1.62
W17	0.17	0.14	0.08	0.24	0.003	0.000 (0.001)	0.65	1.29
W18	0.07	0.11	1.24	0.30	0.003	0.000 (0.001)	1.48	3.20
W19	0.07	0.72	0.12	0.20	0.001 (0.001)	0.000 (0.001)	0.66	1.76
W21	0.13	0.32	0.08	0.16	0.006	0.002	0.55	1.25
W22	0.13	0.53	0.09	0.24	0.018	0.003	0.46	1.48
W23	0.13	0.40	0.14	0.33	0.022	0.003	0.68	1.70
W24	0.40	0.40	0.20	0.49	0.009	0.001 (0.001)	0.68	2.18
W25	0.75	0.52	0.18	0.37	0.020	0.004	0.59	2.44
W26	0.47	0.46	0.04	0.03	0.001 (0.001)	0.000 (0.001)	0.39	1.40
W27	0.55	0.38	0.12	0.21	0.007	0.003	0.75	2.02
W28	0.18	0.14	0.08	0.10	0.005	0.002	0.62	1.12
W29	0.20	0.17	0.07	0.11	0.001 (0.001)	0.002	0.91	1.46
W30	0.10	0.15	0.06	0.13	0.004	0.012	0.66	1.12
W31	0.15	0.13	0.06	0.12	0.001 (0.001)	0.001 (0.001)	0.38	0.83
W32	0.35	0.15	0.10	0.16	0.005	0.001 (0.001)	0.49	1.26
W33	1.12	0.41	0.41	0.66	0.020	0.000 (0.001)	1.84	4.46
W34	0.37	0.19	0.19	0.27	0.002	0.000 (0.001)	0.89	1.91
W35	0.15	0.31	0.21	0.22	0.049	0.005	1.28	2.23
W36	0.47	0.56	0.49	0.58	0.007	0.007	2.32	4.44
W37	2.14	0.54	0.48	0.71	0.003	0.007	1.37	5.26
Mean	0.48	0.38	0.19	0.37	0.012	0.002	1.19	2.62
SD	0.51	0.24	0.21	0.30	0.019	0.003	0.83	1.52

Median	0.22	0.34	0.12	0.24	0.005	0.001 (0.001)	0.90	2.09
Min	0.07	0.11	0.04	0.03	0.001 (0.001)	0.000 (0.001)	0.38	0.83
Max	2.14	0.98	1.24	1.38	0.110	0.013	3.52	6.88
PAE in particle phase (C_{spm}, ng/L)								
W01	0.046	0.11	0.05 (0.13)	0.18	0.010	0.000 (0.002)	1.58	1.97 (1.06)
W02	0.030	0.09	0.07 (0.10)	0.20	0.011	0.000 (0.001)	0.92	1.32
W03	0.024	0.08	0.05 (0.08)	0.11	0.015	0.000 (0.001)	0.99	1.27
W04	0.030	0.08	0.03 (0.07)	0.07	0.008	0.000 (0.001)	2.28	2.50
W05	0.012	0.05	0.03 (0.07)	0.07	0.005	0.000 (0.001)	0.74	0.91
W06	0.038	0.08	0.05 (0.07)	0.16	0.009	0.000 (0.001)	0.64	0.98
W07	0.021	0.06	0.03 (0.05)	0.11	0.006	0.000 (0.001)	0.49	0.73
W08	0.019	0.05	0.02 (0.07)	0.23	0.026	0.002	0.57	0.92
W09	0.072	0.20 (0.21)	0.13 (0.36)	0.43	0.013	0.000 (0.005)	2.1 (2.2)	2.94
W10	0.018	0.04 (0.045)	0.11	0.11	0.015	0.001 (0.001)	0.39 (0.47)	0.69
W11	0.037	0.09 (0.09)	0.23	0.28	0.027	0.002 (0.002)	0.74 (0.95)	1.41
W12	0.017	0.04 (0.06)	0.02 (0.11)	0.03	0.018	0.003	0.43 (0.67)	0.57 (0.89)
W13	0.047	0.09 (0.11)	0.1 (0.19)	0.06	0.024	0.003	0.57(1.17)	0.91 (1.55)
W14	0.040	0.08	0.03 (0.12)	0.04	0.002 (0.003)	0.000 (0.002)	0.44 (0.76)	0.63 (1.0)
W15	0.007	0.02 (0.02)	0.01 (0.04)	0.01	0.001 (0.001)	0.000 (0.001)	0.12 (0.26)	0.17 (0.34)
W16	0.035	0.05	0.02 (0.07)	0.04	0.002 (0.002)	0.000 (0.001)	0.28 (0.40)	0.43 (0.53)
W17	0.038	0.06	0.01 (0.07)	0.08	0.001 (0.002)	0.000 (0.001)	0.28 (0.40)	0.47 (0.53)
W18	0.012 (0.015)	0.03 (0.06)	0.03 (0.10)	0.06	0.001 (0.003)	0.000 (0.001)	0.18 (0.63)	0.31 (0.84)
W19	0.003 (0.008)	0.01 (0.03)	0.02 (0.05)	0.01 (0.01)	0.000 (0.001)	0.000 (0.001)	0.12 (0.32)	0.16 (0.43)
W21	0.007 (0.010)	0.10	0.07 (0.07)	0.14	0.020	0.000 (0.001)	0.94	1.27
W22	0.008 (0.010)	0.09	0.09	0.13	0.017	0.000 (0.001)	0.61	0.95
W23	0.011 (0.010)	0.11	0.06 (0.08)	0.10	0.013	0.000 (0.001)	0.6	0.89
W24	0.011 (0.010)	0.10	0.06 (0.08)	0.10	0.013	0.000 (0.001)	0.61	0.89
W25	0.009 (0.009)	0.09	0.06 (0.06)	0.09	0.011	0.000 (0.001)	0.59	0.84
W26	0.004 (0.009)	0.06	0.04 (0.06)	0.08	0.009	0.000 (0.001)	0.33 (0.38)	0.52
W27	0.009 (0.013)	0.09	0.04 (0.09)	0.06	0.005	0.000 (0.001)	0.39 (0.56)	0.59 (0.74)
W28	0.012 (0.012)	0.08	0.04 (0.08)	0.06	0.006	0.000 (0.001)	0.33 (0.50)	0.53
W29	0.011 (0.010)	0.08	0.04 (0.07)	0.07	0.006	0.000 (0.001)	0.37 (0.43)	0.59
W30	0.005 (0.007)	0.05	0.03 (0.05)	0.04	0.004	0.000 (0.001)	0.43	0.55
W31	0.006 (0.008)	0.03 (0.03)	0.02 (0.05)	0.03	0.002	0.000 (0.001)	0.16 (0.33)	0.24 (0.44)
W32	0.004 (0.008)	0.04	0.02 (0.06)	0.03	0.003	0.000 (0.001)	0.19 (0.36)	0.28 (0.48)
W33	0.005 (0.007)	0.03 (0.03)	0.02 (0.05)	0.02	0.001 (0.001)	0.000 (0.001)	0.53	0.60
W34	0.013 (0.014)	0.06	0.03 (0.10)	0.04	0.003	0.000 (0.001)	0.26 (0.58)	0.41 (0.77)
W35	0.009 (0.009)	0.06	0.02 (0.07)	0.04	0.002 (0.002)	0.000 (0.001)	0.2 (0.44)	0.33 (0.59)
W36	0.190	0.28	0.15 (0.37)	0.15	0.004 (0.01)	0.000 (0.005)	1.01 (2.28)	1.79 (3.02)
W37	0.034	0.05 (0.05)	0.05 (0.13)	0.05	0.005	0.000 (0.002)	0.27 (0.80)	0.46 (1.06)
Mean	0.025	0.076	0.052 (0.077)	0.098	0.009	0.000 (0.001)	0.60	0.86
SD	0.032	0.049	0.045 (0.077)	0.085	0.007	0.001 (0.001)	0.49	0.62 (0.62)
Median	0.013	0.070	0.040 (0.077)	0.074	0.006	0.000 (0.001)	0.47 (0.47)	0.66

Min	0.003 (0.011)	0.013	0.009 (0.077)	0.011 (0.017)	0.000 (0.002)	0.000 (0.001)	0.12 (0.47)	0.16 (0.62)
Max	0.190	0.279	0.229	0.428	0.027	0.003	2.27	2.94
Sum of $C_W + C_{SPM}$ (ng/L)								
W01	0.24	0.57	0.30	1.26	0.012	0.000 (0.003)	3.11	5.51
W02	0.18	0.36	0.27	1.58	0.009	0.000 (0.003)	2.31	4.73
W03	0.43	0.38	0.20	0.84	0.010	0.001 (0.002)	1.76	3.63
W04	0.15	0.22	0.21	0.56	0.006	0.000 (0.002)	3.20	4.35
W05	0.21	0.30	0.12	0.37	0.005	0.000 (0.002)	2.06	3.07
W06	0.28	0.23	0.16	0.67	0.006	0.001 (0.002)	1.28	2.62
W07	0.15	0.32	0.12	0.39	0.005	0.000 (0.001)	1.47	2.47
W08	1.62	0.98	0.33	0.44	0.016	0.008	1.05	4.47
W09	1.21	1.04	0.38	1.44	0.060	0.000 (0.009)	5.62	9.82
W10	1.32	0.61	0.24	0.28	0.021	0.004	2.42	4.92
W11	1.20	0.85	0.43	0.71	0.020	0.002 (0.004)	3.42	6.65
W12	0.79	0.40	0.09	0.23	0.011	0.002 (0.003)	1.45	2.99
W13	1.34	1.08	0.18	0.26	0.014	0.002 (0.005)	3.44	6.34
W14	0.29	0.51	0.12	0.26	0.005 (0.005)	0.000 (0.003)	3.74	4.93
W15	0.14	0.23	0.06	0.14	0.002 (0.002)	0.000 (0.001)	0.82	1.40
W16	0.17	0.24	0.09	0.21	0.011	0.000 (0.002)	1.32	2.06
W17	0.21	0.20	0.10	0.32	0.002 (0.003)	0.000 (0.002)	0.93	1.76
W18	0.08	0.14	1.27	0.36	0.002 (0.004)	0.000 (0.003)	1.66	3.51
W19	0.07	0.74	0.13	0.21	0.001 (0.002)	0.000 (0.001)	0.78	1.93
W21	0.14	0.42	0.15	0.30	0.013	0.001 (0.002)	1.49	2.52
W22	0.14	0.62	0.18	0.38	0.017	0.002 (0.002)	1.07	2.43
W23	0.14	0.51	0.20	0.43	0.017	0.001 (0.002)	1.27	2.59
W24	0.41	0.50	0.26	0.59	0.011	0.001 (0.002)	1.29	3.07
W25	0.76	0.61	0.24	0.46	0.016	0.002 (0.002)	1.18	3.28
W26	0.47	0.52	0.09	0.11	0.005	0.000 (0.002)	0.72	1.92
W27	0.56	0.47	0.16	0.26	0.006	0.002 (0.002)	1.14	2.61
W28	0.20	0.21	0.12	0.15	0.005	0.001 (0.002)	0.95	1.65
W29	0.21	0.25	0.11	0.19	0.004	0.001 (0.002)	1.28	2.05
W30	0.10	0.20	0.09	0.18	0.004	0.006	1.08	1.67
W31	0.16	0.16	0.08	0.15	0.001 (0.002)	0.000 (0.001)	0.53	1.08
W32	0.36	0.19	0.12	0.19	0.004	0.001 (0.002)	0.68	1.54
W33	1.13	0.44	0.43	0.68	0.011	0.000 (0.001)	2.36	5.06
W34	0.38	0.25	0.22	0.31	0.003 (0.004)	0.000 (0.002)	1.15	2.32
W35	0.16	0.37	0.23	0.26	0.025	0.002 (0.002)	1.48	2.56
W36	0.66	0.84	0.64	0.74	0.005 (0.015)	0.004 (0.010)	3.33	6.23
W37	2.17	0.59	0.53	0.76	0.004 (0.005)	0.004	1.64	5.72
Mean	0.51	0.46	0.24	0.46	0.02	0.003 (0.002)	1.79	3.48
SD	0.51	0.26	0.22	0.36	0.02	0.003 (0.002)	1.11	1.89
Median	0.26	0.41	0.18	0.34	0.01	0.000 (0.002)	1.38	2.81
Min	0.07	0.14	0.06	0.11	0.002 (0.003)	0.000 (0.002)	0.53	1.08

Max	2.17	1.08	1.27	1.58	0.12	0.02	5.62	9.82

Table S 12. Comparison with previous studies for the concentrations of seven PAEs in seawater (ng/L)

Region	DMP	DEP	DiBP	DnBP	BBP	DCHP	DEHP
Arctic[11]	n.d.- 0.3	n.d.-0.8	n.d.-0.2	0.008-0.3	n.d.-0.05	-	n.d.-3.3
North Sea (Germany)[5]	0.23	1.45	-	1.74	0.07	-	3.8
North Sea (Belgium)[12]	-	< 25-753	-	< 5-2645	< 10-343	-	66-766
East China Sea, China[13-15]	0.08-830	0.25-2404	5-17256	11-17952	0.09-344	0.05-705	9- 9738
Bohai and Yellow Seas, China[16, 17]	n.d.-160	1.18-85.9	89.3-2424	n.d.-2226	n.d.-7.8	1.34-3.12	51.4-3388
Pearl River Delta, China[18]	1.7-13.6	＜5.1-34.8	＜30.2-70.3	＜33.2-358	n.d.	n.d.	12.7-1050
Mediterranean Sea[19-21]	1.4-140	6.9-870	56.5-383.4	63.4-466	1-100	-	30-5970
Bay of Biscay, Spain[22]	7.5±0.4	33±3	-	83±7	8±1	64±4	-
Barkley Sound, Canada[23]	-	-	-	18-3000	-	-	10-950
Puget Sound, USA[23]	-	-	-	-	-	-	60-640
Klang River estuary, Australia[24]	-	-	-	-	-	-	3100-64300
South Korea[25]	20-100	20-150	-	40-360	-	-	30-300
Tropical western Pacific Ocean[26]	n.d.-7	n.d.-2.1	1.9-14	2.2-13	n.d.-5.5	1.1-7	2.0-9.2
Caspian Sea, Iran[27]	490	520	-	-	-	-	-
Thailand[28]	-	-	-	230-770	-	-	310-1160
Tunisia[29]	-	< 10-17000	< 5-106000	< 29-30500	-	-	< 26-168000
This work	0.07-2.17	0.14-1.08	0.06-1.27	0.11-1.58	<n.d.-0.12	<n.d.-0.02	0.53-5.62

Table S 13. Particle-bound fractions (ø) of PAEs in seawater from the South China Sea

Sample	DMP	DEP	DiBP	DnBP	DEHP	PAE
W01	0.19	0.20	0.16	0.14	0.51	1.80
W02	0.16	0.26	0.24	0.13	0.40	1.90
W03	0.06	0.21	0.27	0.13	0.56	2.11
W04	0.20	0.35	0.15	0.13	0.71	2.30
W05	0.06	0.18	0.23	0.20	0.36	1.71
W06	0.14	0.36	0.31	0.24	0.50	2.48
W07	0.13	0.19	0.29	0.29	0.33	2.03
W08	0.01	0.05	0.07	0.52	0.55	2.14
W09	0.06	0.19	0.33	0.30	0.37	1.67
W10	0.01	0.07	0.46	0.39	0.16	1.54
W11	0.03	0.10	0.54	0.40	0.22	2.71
W12	0.02	0.11	0.18	0.15	0.30	2.48
W13	0.04	0.09	0.57	0.23	0.17	2.82
W14	0.14	0.15	0.21	0.16	0.12	1.26
W15	0.05	0.08	0.15	0.08	0.15	0.80
W16	0.20	0.22	0.21	0.21	0.21	1.19
W17	0.18	0.32	0.15	0.24	0.30	1.62
W18	0.15	0.24	0.02	0.16	0.11	0.94
W19	0.04	0.02	0.11	0.06	0.16	0.96
W21	0.05	0.23	0.47	0.45	0.63	2.65
W22	0.06	0.15	0.48	0.36	0.57	2.14
W23	0.07	0.21	0.29	0.24	0.47	1.69
W24	0.02	0.20	0.22	0.18	0.47	1.72
W25	0.01	0.14	0.24	0.19	0.50	1.51
W26	0.01	0.12	0.49	0.70	0.45	3.07
W27	0.02	0.19	0.26	0.22	0.34	1.50
W28	0.06	0.36	0.32	0.38	0.35	2.08
W29	0.05	0.33	0.38	0.39	0.29	2.30
W30	0.05	0.23	0.28	0.25	0.40	1.74
W31	0.04	0.20	0.23	0.18	0.29	1.80
W32	0.01	0.20	0.17	0.15	0.28	1.21
W33	0.00	0.06	0.04	0.03	0.22	0.56
W34	0.04	0.23	0.14	0.14	0.22	1.37
W35	0.05	0.15	0.09	0.14	0.14	0.62
W36	0.29	0.33	0.23	0.21	0.30	1.74
W37	0.02	0.09	0.10	0.06	0.17	1.11
Mean	0.08	0.19	0.25	0.23	0.34	1.76
SD	0.07	0.09	0.14	0.14	0.16	0.62

Table S 14. Correlation analysis of PAEs in air

	DMP	DEP	DiBP	DnBP	DEHP
DMP	1(0.000***)	0.647(0.001***)	0.242(0.279)	0.168(0.455)	0.214(0.339)
DEP	0.647(0.001***)	1(0.000***)	0.385(0.077*)	0.398(0.067*)	0.199(0.375)
DiBP	0.242(0.279)	0.385(0.077*)	1(0.000***)	0.951(0.000***)	-0.091(0.686)
DnBP	0.168(0.455)	0.398(0.067*)	0.951(0.000***)	1(0.000***)	-0.148(0.512)
DEHP	0.214(0.339)	0.199(0.375)	-0.091(0.686)	-0.148(0.512)	1(0.000***)

***, **, *means significant 0.01, 0.05, 0.1

Table S 15. Correlation analysis of PAEs in seawater

	DMP	DEP	DiBP	DnBP	DEHP
DMP	1(0.000***)	0.659(0.000***)	0.27(0.112)	0.245(0.150)	0.354(0.034**)
DEP	0.659(0.000***)	1(0.000***)	0.174(0.311)	0.336(0.045**)	0.55(0.001***)
DiBP	0.27(0.112)	0.174(0.311)	1(0.000***)	0.324(0.054*)	0.296(0.080*)
DnBP	0.245(0.150)	0.336(0.045**)	0.324(0.054*)	1(0.000***)	0.611(0.000***)
DEHP	0.354(0.034**)	0.55(0.001***)	0.296(0.080*)	0.611(0.000***)	1(0.000***)

***, **,*means significant 0.01, 0.05, 0.1

Table S 16. Air-sea exchange fluxes of PAEs (DMP, DEP, DiBP, DnBP and DEHP) in the South China Sea. Positive value indicates water to air volatilization and negative value indicates air to water deposition

F ($ng/m^2/day$)	DMP	DEP	DiBP	DnBP	DEHP
W01	-970	-85	-1488	-1015	918
W02	-888	-71	-657	-435	57
W03	-969	-88	-1502	-1048	419
W04	-391	-79	-1278	-899	331
W05	-392	-78	-1337	-947	919
W06	-243	-147	-671	-489	314
W07	-243	-146	-674	-505	528
W08	-241	-109	-440	-444	470
W09	-245	-110	-434	-335	3538
W10	-244	-118	-457	-445	2255
W11	-174	-79	-303	-279	1419
W12	-176	-86	-313	-297	492
W13	-210	-102	-1054	-874	1272
W14	-217	-112	-1138	-943	3616
W15	-104	-41	-172	-198	733
W16	-106	-42	-172	-197	1059
W17	-244	-94	-678	-465	444
W18	-258	-100	-628	-494	1593
W19	-258	-90	-726	-505	601
W21	-164	-53	-107	-48	740
W22	-181	-161	-959	-634	338
W23	-181	-163	-961	-631	600
W24	-177	-88	-2080	-1470	310
W25	-174	-86	-2042	-1451	214
W26	-177	-87	-2178	-1568	233
W27	-460	-104	-1349	-1050	435
W28	-419	-92	-1120	-927	33
W29	-442	-104	-1896	-1579	255
W30	-443	-105	-1948	-1622	194
W31	-279	-42	-284	-230	159
W32	-371	-42	-12	-40	273
W33	-1487	-383	-1162	-1500	1308
W34	-1497	-391	-1226	-1596	663
W35	-44	-21	-328	-229	329
W36	-44	-19	-342	-231	1366
W37	-190	-77	-406	-326	361
mean	-370	-105	-903	-721	800

Table S 17. Particle dry deposition of PAEs in the South China Sea

F_D (ng/m^2/day)	DMP	DEP	DiBP	DnBP	DEHP
A01	1.2	3.6	2.2	2.8	94
A02	0.9	2.2	16.5	10.9	198
A03	15.4	35.7	1396	918	179
A04	4.7	12.4	1097	623	85
A05	2.8	14.6	877	473	104
A06	3.4	6.7	508	590	67
A07	1.9	6.5	63	284	68
A08	1.8	7.0	165	214	54
A09	3.8	12.2	496	454	67
A10	2.7	8.0	1378	1135	126
A11	2.4	8.3	528	612	92
A12	1.4	2.8	53	57	40
A13	4.4	5.9	62	83	76
A14	3.1	12.5	17	41	86
A15	3.5	24.4	553	411	80
A16	28.1	24.4	22	29	96
A17	2.1	11.0	25	40	103
A18	5.0	22.4	53	54	118
A19	1.3	3.1	1.6	4.6	26
A20	4.7	11.4	634	430	55
A21	6.8	34.7	1814	1631	138
A22	2.2	5.2	6.4	10	58
Max	28	35.7	1814	1631	198
Min	0.86	2.21	1.6	2.8	26
Mean	4.71	12.5	444	368	91
SD	6.04	9.81	548	427	42
Median	2.93	9.68	114	249	86

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