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Secondary formation dominated low molecular weight amines origins in aerosols over the marginal seas of China

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Supplementary tables

Table S1. Summary of sampling information during the cruise.

Sample ID	Sampling period		Sampling duration (min)	Midpoint position of the sampling period		Sea area	Average ambient temperature (°C)	Average relative humidity (%)	Average wind speed (m s^{-1})
	Start time	End time		Longitude (°E)	Latitude (°N)				
S2	16:30, 28 Mar.	16:30, 29 Mar.	1020	122.32	36.00	SYS	8.2	99.3	6.4
S3	16:38, 29 Mar.	16:20, 30 Mar.	810	123.95	34.98	SYS	9.1	86.3	5.2
S4	17:20, 30 Mar.	15:10, 31 Mar.	1170	121.12	34.67	SYS	8.8	99.2	4.9
S5	15:40, 31 Mar.	15:13, 1 Apr.	1110	124.00	34.00	SYS	12.2	100.0	6.9
S6	15:15, 1 Apr.	15:20, 2 Apr.	1050	122.15	32.65	SYS	12.1	100.0	7.2
S8	15:30, 2 Apr.	20:40, 2 Apr.	450	123.98	32.00	SYS	13.0	99.7	6.6
S10	15:30, 3 Apr.	19:34, 3 Apr.	450	122.23	35.50	SYS	7.8	100.0	11.1
S12	16:30, 7 Apr.	17:41, 8 Apr.	840	123.70	37.15	SYS	6.2	68.9	6.7
S13	19:11, 8 Apr.	18:20, 9 Apr.	1050	121.85	38.63	NYS	7.1	83.4	6.7
S14	09:00, 10 Apr.	07:50, 11 Apr.	810	119.38	37.80	BS	10.9	67.4	7.7
S15	16:53, 11 Apr.	16:53, 12 Apr.	630	119.93	39.22	BS	7.8	67.4	7.5
S16	18:03, 12 Apr.	18:00, 13 Apr.	540	120.63	38.33	BS	8.3	66.6	6.6
S17	18:05, 13 Apr.	20:00, 14 Apr.	1410	123.12	38.75	NYS	6.1	93.2	5.0
S18	14:31, 15 Apr.	14:33, 16 Apr.	810	123.08	38.15	NYS	7.1	79.8	4.7
S19	15:00, 16 Apr.	23:58, 16 Apr.	540	121.78	36.35	SYS	9.1	81.5	5.2

Table S2. Statistical summary of measured chemical components.

Component (ng m^{-3})	SYS (N = 9)	NYS (N = 3)	BS (N = 3)
Amines	40.4 ± 16.4	43.5 ± 17.5	63.6 ± 18.3
MA	10.0 ± 7.0	15.7 ± 7.7	22.8 ± 15.0
EA	1.7 ± 0.6	2.0 ± 1.8	3.0 ± 1.3
DMA	3.5 ± 2.1	3.8 ± 2.6	7.9 ± 2.1
IPA	1.9 ± 0.9	1.3 ± 0.7	2.1 ± 1.4
PA	2.5 ± 0.9	3.0 ± 0.7	4.1 ± 0.7
TMDEA	20.7 ± 9.1	17.8 ± 7.3	23.8 ± 3.7
Water soluble inorganic ions (WSIIs)	8692.1 ± 4363.1	13043.1 ± 6299.9	26912.3 ± 4926.0
Na^+	510.4 ± 455.1	714.7 ± 459.3	1626.9 ± 323.8
NH_4^+	1642.1 ± 657.5	2357.2 ± 989.6	3778.2 ± 481.2
K^+	162.8 ± 70.5	183.3 ± 74.7	481.4 ± 110.4
Mg^{2+}	41.3 ± 38.9	82.9 ± 51.3	271.0 ± 49.4
Ca^{2+}	253.5 ± 199.1	456.6 ± 199.6	1949.9 ± 441.7
F^-	12.3 ± 10.1	28.6 ± 31.2	36.1 ± 7.3
Cl^-	237.6 ± 411.3	602.2 ± 425.1	1582.1 ± 570.7
NO_2^-	4.4 ± 3.5	2.8 ± 2.2	4.3 ± 5.3
NO_3^-	1805.9 ± 2097.6	5255.1 ± 2655.6	10569.0 ± 3189.7
SO_4^{2-}	4021.8 ± 2178.3	3359.6 ± 1544.8	6613.4 ± 893.5
Low molecular organic acids	249.3 ± 124.3	204.0 ± 91.0	247.7 ± 17.0
CHO_2^-	31.8 ± 8.9	38.8 ± 26.2	50.6 ± 10.6
$C_2H_3O_2^-$	29.5 ± 13.5	27.4 ± 10.8	52.1 ± 20.1
$C_4H_4O_4^{2-}$	44.9 ± 36.0	40.5 ± 28.1	58.2 ± 8.3
$C_5H_6O_4^{2-}$	43.7 ± 26.3	47.9 ± 27.4	47.1 ± 2.2
$CH_3O_3S^-$ (MSA^-)	99.5 ± 76.1	49.5 ± 39.9	39.7 ± 8.2
Carbonaceous components (TC)	4670.7 ± 1511.5	5911.3 ± 1775.7	9467.5 ± 1774.8
Organic carbon (OC)	4227.8 ± 1375.7	5126.6 ± 1344.7	8448.9 ± 1627.7
Elemental carbon (EC)	442.9 ± 159.5	784.7 ±431.6	1018.6 ± 156.8
Organic compositions	126.2 ± 74.3	230.6 ± 159.8	474.8 ± 174.6
n-Alkanes (ALK, C_{14}–C_{35})	41.4 ± 22.5	50.6 ± 20.8	93.8 ± 29.1
ALK_{LMW} (C_{20}–C_{26})	22.9 ± 13.8	25.4 ± 10.3	40.3 ± 10.6
ALK_{HMW} (C_{27}, C_{29}, C_{31}, C_{33})	11.2 ± 6.0	16.9 ± 7.4	35.9 ± 13.5
Fatty acids (FA, $C_{14:0}$–$C_{30:0}$)	33.4 ± 15.4	56.4 ± 49.9	67.9 ± 25.3
FA_{LMW} ($\leq C_{19:0}$)	24.1 ± 6.5	29.8 ± 19.6	54.0 ± 20.1
FA_{HMW} ($> C_{19:0}$)	3.6 ± 3.0	10.3 ± 8.3	11.5 ± 4.1
Fatty alcohols (ALC, C_{15}–C_{30})	2.9 ± 1.3	5.0 ± 3.0	8.7 ± 3.2
ALC_{LMW} ($\leq C_{19alc}$)	1.4 ± 0.5	1.0 ± 0.6	1.5 ± 0.4

Component (ng m^{-3})	SYS (N = 9)	NYS (N = 3)	BS (N = 3)
ALC_{HMW} (> C_{19alc})	1.5 ± 0.9	3.9 ± 2.4	7.2 ± 3.0
Polycyclic aromatic hydrocarbons (PAHs)	0.9 ± 0.8	2.0 ± 1.1	3.9 ± 1.5
Hopanes	3.0 ± 3.7	2.4 ± 1.3	4.6 ± 2.5
Steranes	0.5 ± 0.5	0.4 ± 0.2	0.9 ± 0.5
Anhydrosugars	20.0 ± 27.4	46.2 ± 36.7	69.8 ± 22.9
Lignin products	2.0 ± 1.5	4.3 ± 2.4	6.6 ± 1.4
Primary sugars and sugar alcohols	11.8 ± 10.7	37.4 ± 32.0	190.0 ± 97.4
Hydroxy-/polyacids	3.4 ± 2.9	11.5 ± 9.1	7.8 ± 1.7
Aromatic acids	3.4 ± 2.3	7.3 ± 4.1	10.0 ± 2.4
Isoprene SOA (SOA_I)	0.4 ± 0.2	0.9 ± 0.6	1.7 ± 0.8
Monoterpene SOA (SOA_M)	2.1 ± 1.2	4.8 ± 2.8	7.6 ± 3.3
β-Caryophyllene SOA (SOA_C)	0.3 ± 0.3	0.5 ± 0.3	0.8 ± 0.3
2,3-dihydroxy-4-oxopentanoic acid (DHOPA)	0.5 ± 0.3	0.8 ± 0.4	0.6 ± 0.1

Note: The data format is mean ± standard deviation.

Table S3. Concentrations of amines in marine and coastal atmospheres.

Sea area	Sampling period	Particle size or gas	Concentrations of amines (ng m^{-3})						Reference
			MA	EA	DMA	IPA	PA	TMDEA	
SYS	March−April 2018	TSP	10.0	1.7	3.5	1.9	2.5	20.7	This study
NYS	April 2018		15.7	2.0	3.8	1.3	3.0	17.8	
BS	April 2018		22.8	3.0	7.9	2.1	4.1	23.8	
ECS−SCS−YS	April−May 2018	Gas			11.0			5.4	(Chen et al., 2022)
(Coastline)	April−May 2018	$PM_{2.5}$			< 4−100*			< 2−21*	
Coastal Qingdao, China	January–February 2018	$PM_{2.5}$	8.5	2.7	58.7		0.9	19.3	(Liu et al., 2022)
	November–December 2019		6.9	2.4	86.3		0.9	14.6	
Huaniao Island, China	January 2013	$PM_{2.5}$	4.8	0.8			2.5		(Huang et al., 2018)
SYS−ECS	March−May 2015	PM_{10}			18.4			22.8	(Yu et al., 2016)
NWPO	April 2015				12.9			13.2	
SYS	August 2015				42.8			45.6	
SYS	November 2013				19.8			37.0	
SYS	November 2012				13.4			30.0	
NYS−BS	August−September 2015				20.7			28.8	
NYS−BS	November 2012				≈ MDL			15.0	
YS−NWPO	April 2015	$PM_{0.056−10}$			12.9			13.2	(Xie et al., 2018)
ECS	June 2016				30.8			12.0	
YS−BS	June−July 2016				50.6			21.0	
YS−BS	August 2015				23.9			18.6	
SYS	November 2013				18.9			31.8	
Coastal Qingdao, China	August 2016				28.5			9.0	

Sea area	Sampling period	Particle size or gas	Concentrations of amines (ng m^{-3})						Reference
			MA	EA	DMA	IPA	PA	TMDEA	
YS−BS	May 2012	$PM_{0.43}$			46.0			108	(Hu et al., 2015)
		PM_{11}			202			432	
YS−ECS	March–April 2017	$PM_{1.8}$			11.9			14.6	(Zhou et al., 2019)
		PM_{10}			13.5			16.6	
Huaniao Island, China	August 2016	$PM_{2.5}$			4.0			8.7	
	November–December 2016	$PM_{1.8}$			10.7			6.0	
	November–December 2016	PM_{10}			15.1			8.4	
	March 2017	$PM_{1.8}$			6.8			2.7	
	March 2017	PM_{10}			11.4			3.1	
	June–July 2017	$PM_{1.8}$			29.0			24.8	
	June–July 2017	PM_{10}			32.2			27.5	
	August–September 2017	$PM_{1.8}$			25.8			25.0	
	August–September 2017	PM_{10}			27.4			26.3	
Arabian Sea	August–October 1994	$PM_{0.9}$	3.2		2.1			0.3	(Gibb et al., 1999)
	November–December 1994		3.7		11.0			0.5	
North Atlantic	June–July 2006	PM_1			14.7			14.3	(Facchini et al., 2008)
Mace Head, Ireland	January–December 2006				4.7			7.6	
East Mediterranean, Greece	2005–2006	PM_1			9.2			< MDL	(Violaki and Mihalopoulos, 2010)
Cape Verde, northeast Tropical Atlantic	May 2007	$PM_{0.14–0.42}$	0.02		0.22			0.06	(Müller et al., 2009)
	June 2007		0.06		0.2			0.08	
	December 2007		0.18		0.57			0.32	
Jeju Island, South Korea	March–April 2001	$PM_{2.5}$	13.5	3.1					(Yang et al., 2004)

Sea area	Sampling period	Particle size or gas	Concentrations of amines (ng m^{-3})						Reference
			MA	EA	DMA	IPA	PA	TMDEA	
Cape Verde, tropical Atlantic Ocean	November 2011 and 2013	Gas	0.8		4.5			< MDL	(Pinxteren et al., 2019)
	November 2011 and 2013	PM_1	0.2		5.6			3.9	
Off the central coast of California, USA	July 2007	PM_1						22.0	(Sorooshian et al., 2009)

Note: * represent ranges; all other data are mean values; blank cells indicate not measured. Some TMDEA values were reported as TMA, as TMA was expected to overwhelm DEA in marine and coastal environments, although they are co-eluted in IC analysis. The SYS, NYS, BS, ECS, SCS and NWPO represent the South Yellow Sea, North Yellow Sea, Bohai Sea, East China Sea, South China Sea and Northwest Pacific Ocean, respectively.

Table S4. Correlation coefficients between amines and organic molecular tracers in TSP over the YS−BS.

Source	Tracer	MA	EA	DMA	IPA	PA	TMDEA
Primary biogenic sources	Primary sugars and sugar alcohols	**0.72****	**0.55***	**0.64****	−0.12	0.45	0.23
	Fungal spore OC	**0.70****	**0.57***	**0.64****	−0.10	0.49	0.18
	Plant debris OC	**0.72****	**0.53***	**0.66****	−0.12	0.47	0.26
Higher plant waxes	ALK$_{HMW}$	**0.60***	0.51	**0.58***	0.06	**0.60***	0.20
	FA$_{HMW}$	0.39	0.11	0.25	0.00	0.50	0.19
	ALC$_{HMW}$	**0.70****	0.49	**0.58***	−0.12	**0.53***	0.25
Marine/microbial sources	FA$_{LMW}$	0.48	0.29	0.35	0.02	**0.53***	0.14
	ALC$_{LMW}$	0.05	−0.04	−0.05	0.03	0.07	0.06
BSOA	Isoprene SOC	**0.73****	0.44	**055***	−0.18	0.42	0.33
	Monoterpene SOC	**0.69****	0.43	0.47	−0.11	0.43	0.34
	β-Caryophyllene SOC	0.24	0.13	0.35	0.27	**0.67****	0.18
Biomass burning	Levogluctosan (Lev)	0.35	0.23	0.36	−0.00	0.44	0.15
	Lev$_{bb}$	0.45	0.33	0.48	0.02	**0.52***	0.17
	Lignin products	0.49	0.30	0.47	0.09	**0.63***	0.28
Fossil fuel combustion	ALK$_{LMW}$	0.33	0.31	0.46	0.15	**0.67****	0.15
	PAHs	**0.57***	0.41	**054***	0.08	**0.63***	0.21
	Hopanes	0.13	−0.08	0.22	0.27	**055***	0.30
	Steranes	0.09	−0.03	0.22	0.27	**057***	0.19
Secondary oxidation products	Aromatic acids	0.44	0.30	0.40	−0.02	**0.63***	0.19
	Hydroxy-/polyacids	0.26	−0.08	0.04	−0.19	0.25	0.20
	DHOPA	0.16	−0.05	−0.12	0.14	0.01	0.47

Note: ** represent $P < 0.01$, and * represent $P < 0.05$.

Supplementary figures

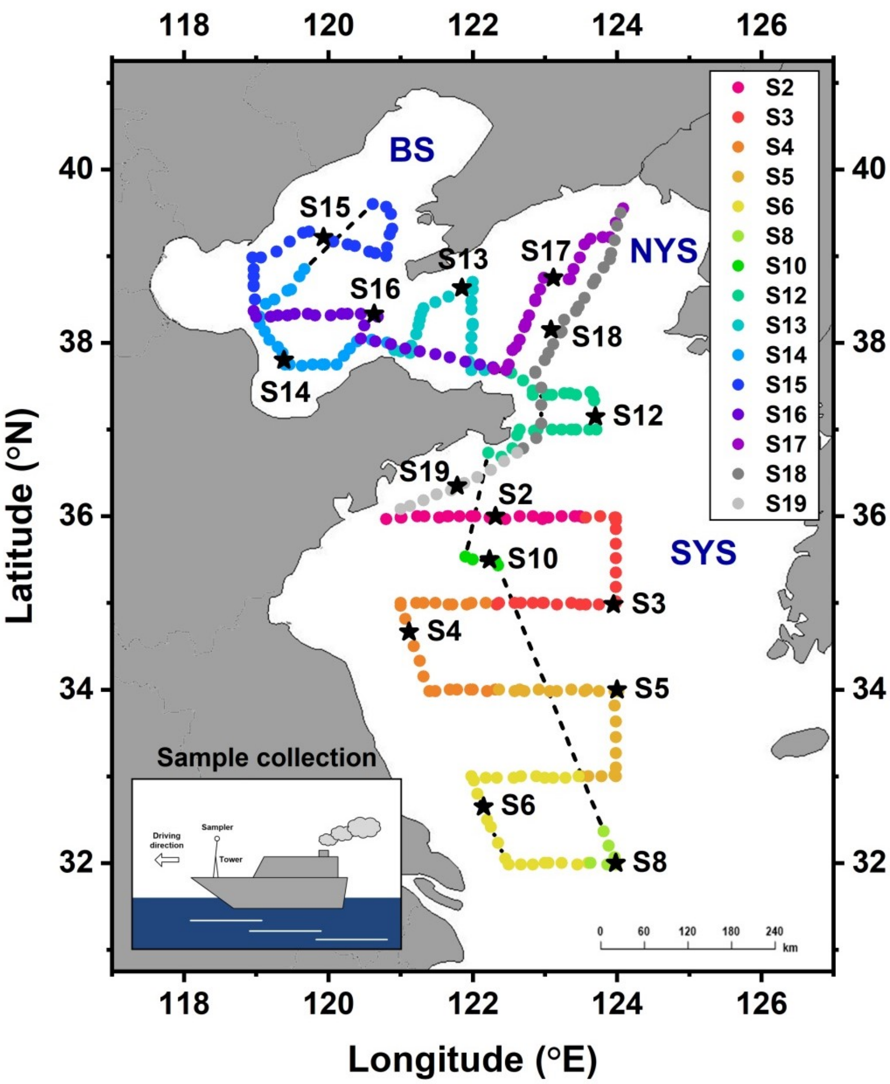

Figure S1. Map of the hourly sampling positions along the cruise track over the YS−BS, together with a schematic diagram of the sampling setup.

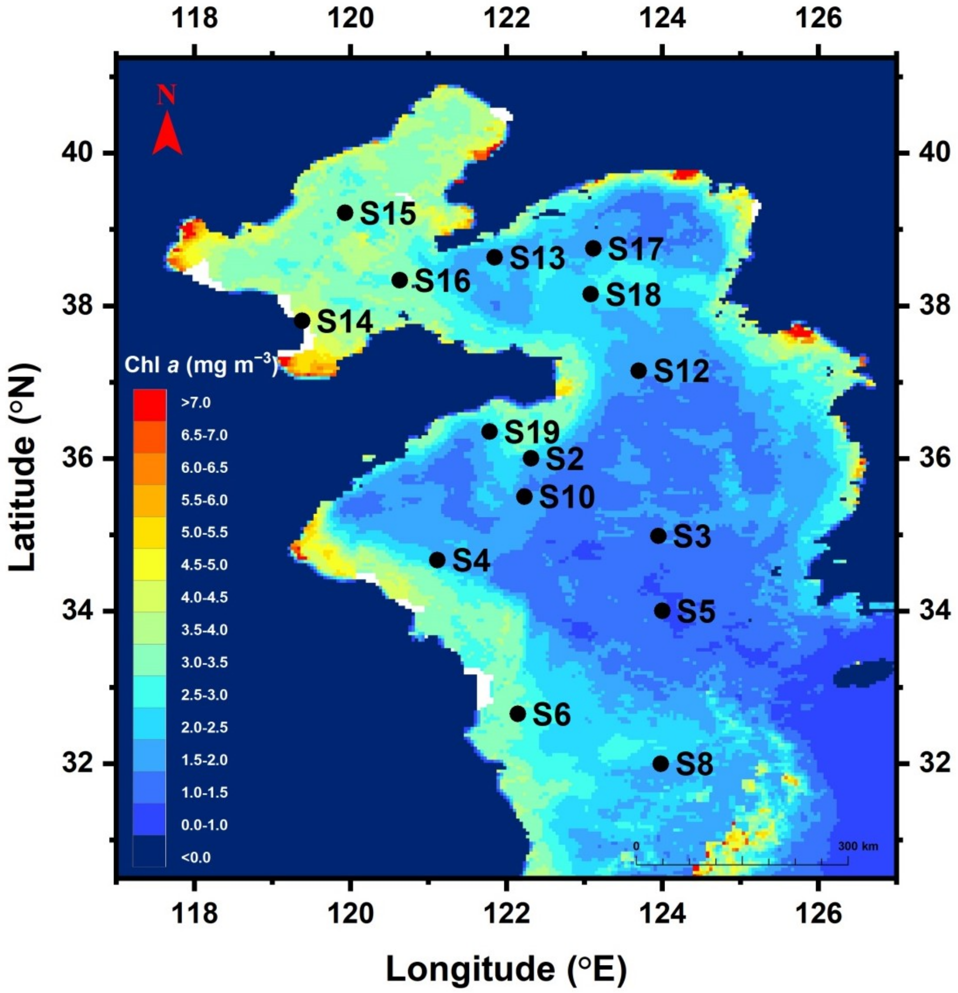

Figure S2. Map of chlorophyll *a* concentrations in surface seawater over the YS−BS during the cruise.

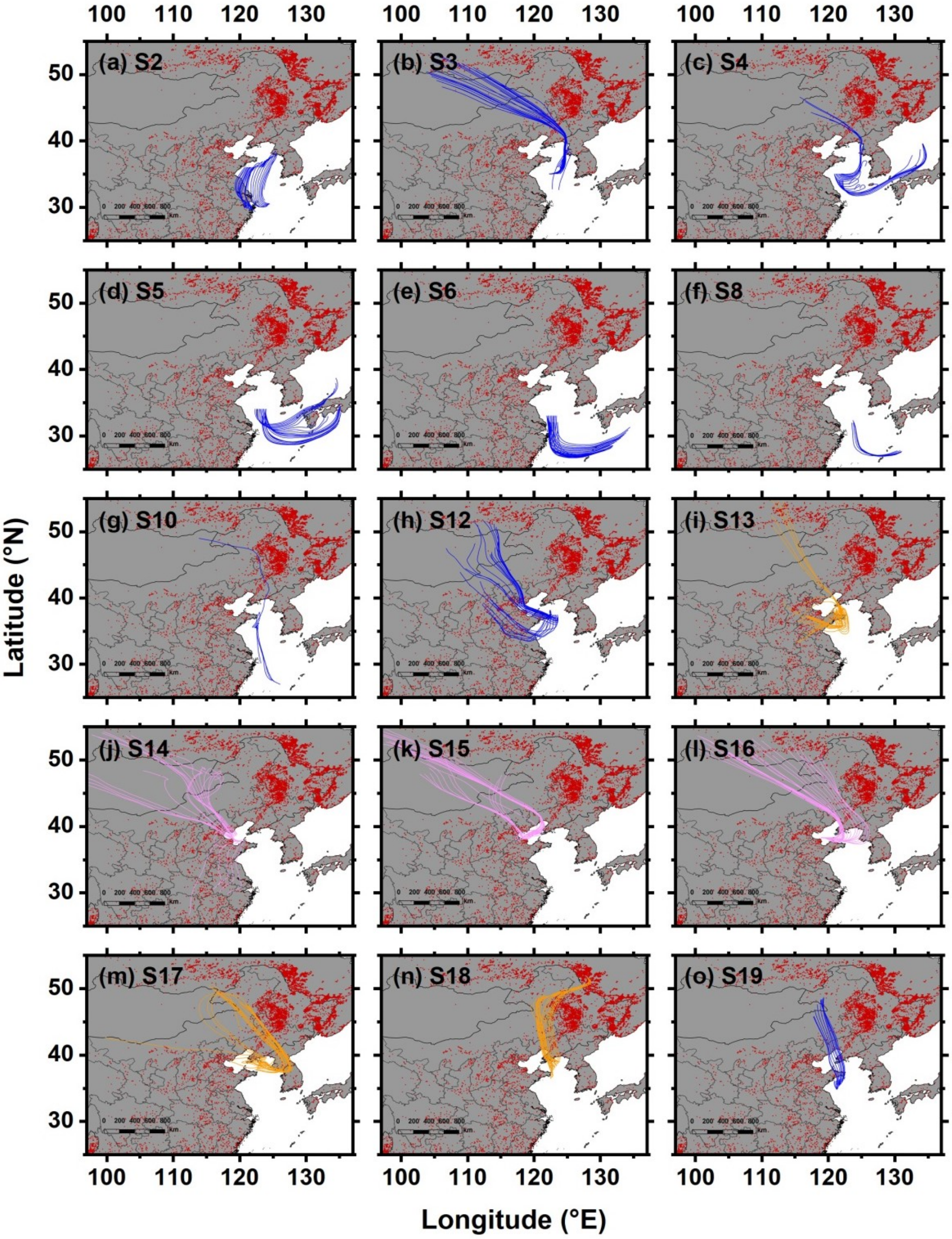

Figure S3. 48 h backward air-mass trajectories starting from the position and time point at the beginning of each sampling, with hourly intervals thereafter. Red dots indicate fire spots during the entire cruise period. Blue, orange, and pink lines represent samples from the SYS, NYS, and BS, respectively.

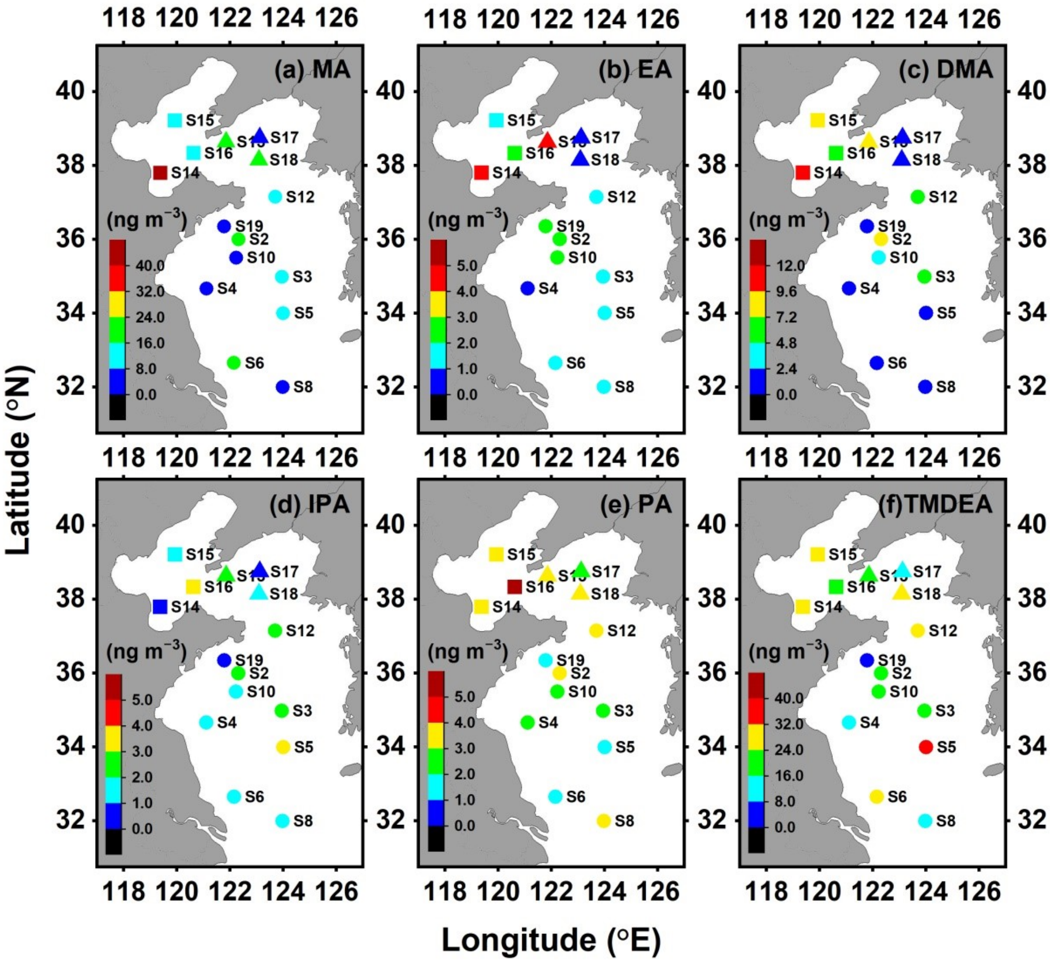

Figure S4. Spatial distributions of MA (a), EA (b), DMA (c), IPA (d), PA (e), and TMDEA (f) in TSP samples over the YS−BS. Circles, triangles, and squares represent samples from the SYS, NYS, and BS, respectively.

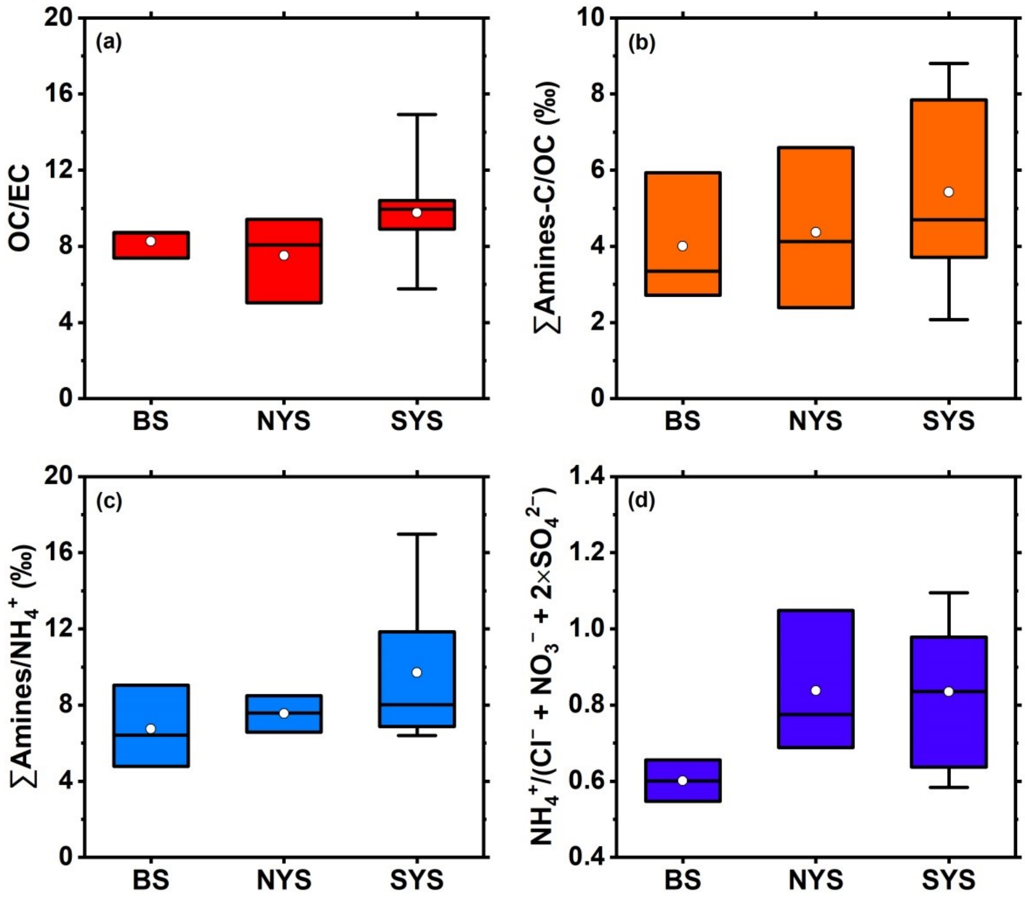

Figure S5. Variations of OC/EC ratios (a), ∑amines-C/OC ratios (b), ∑amines/NH_4^+ molar ratios (c), and $NH_4^+/(Cl^- + NO_3^- + 2\times SO_4^{2-})$ molar ratios (d) in TSP over the SYS, NYS, and BS. Lower and upper box boundaries indicate the 25th and 75th percentiles; lines within the box indicate the medians; dots within the box indicate the mean values; and whiskers below and above the box indicate the minimum and maximum values.

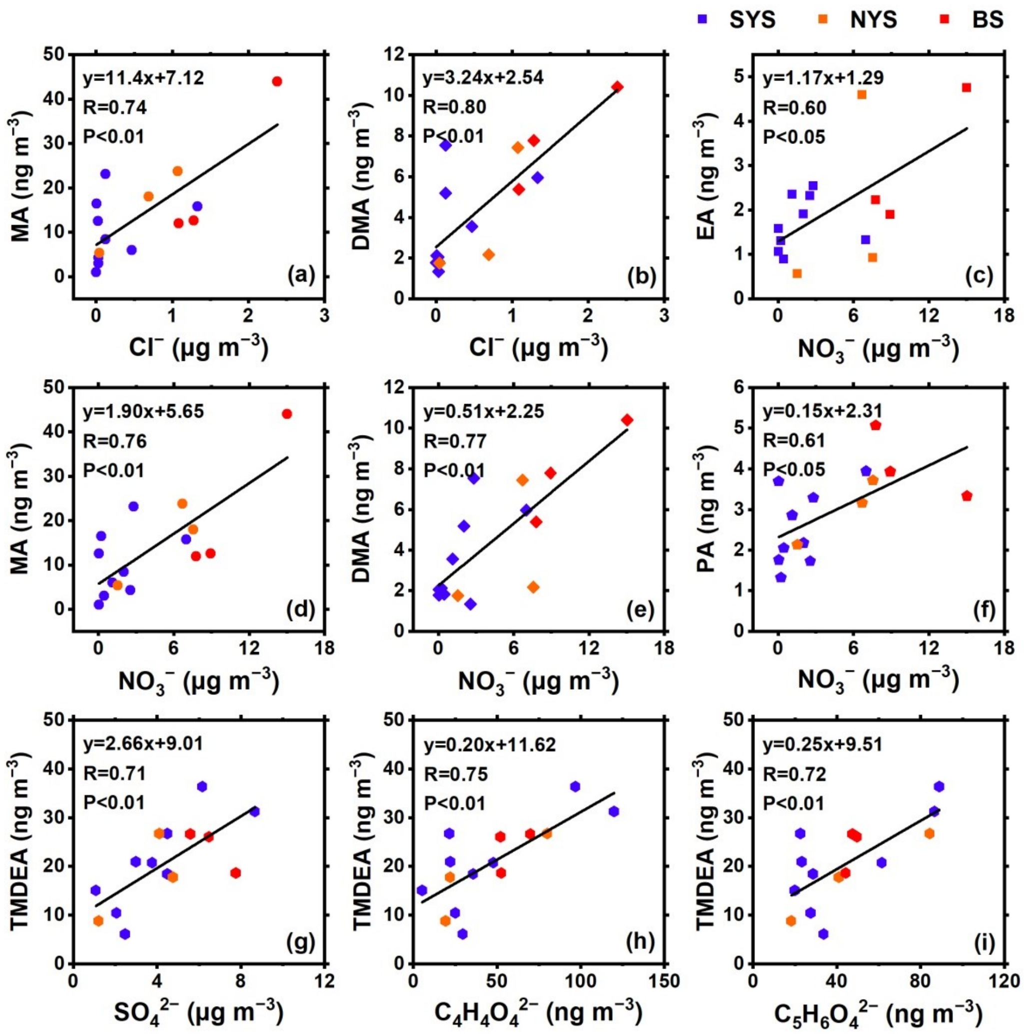

Figure S6. Linear regressions between amines and acidic species in TSP over the SYS, NYS, and BS.

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