Supporting Information

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

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Table S1. Summary of sampling information during the cruise.

Sample ID	Sampling period		Midpoint position of the sampling period		Sea area	Average ambient temperature (℃)	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.	122.32	36.00	SYS	8.2	99.3	6.4
S3	16:38, 29 Mar.	16:20, 30 Mar.	123.95	34.98	SYS	9.1	86.3	5.2
S4	17:20, 30 Mar.	15:10, 31 Mar.	121.12	34.67	SYS	8.8	99.2	4.9
S5	15:40, 31 Mar.	15:13, 1 Apr.	124.00	34.00	SYS	12.2	100.1	6.9
S6	15:15, 1 Apr.	15:20, 2 Apr.	122.15	32.65	SYS	12.1	100.3	7.2
S8	15:30, 2 Apr.	20:40, 2 Apr.	123.98	32.00	SYS	13.0	99.7	6.6
S10	15:30, 3 Apr.	19:34, 3 Apr.	122.23	35.50	SYS	7.8	100.3	11.1
S12	16:30, 7 Apr.	17:41, 8 Apr.	123.70	37.15	SYS	6.2	68.9	6.7
S13	19:11, 8 Apr.	18:20, 9 Apr.	121.85	38.63	NYS	7.1	83.4	6.7
S14	09:00, 10 Apr.	07:50, 11 Apr.	119.38	37.80	BS	10.9	67.4	7.7
S15	16:53, 11 Apr.	16:53, 12 Apr.	119.93	39.22	BS	7.8	67.4	7.5
S16	18:03, 12 Apr.	18:00, 13 Apr.	120.63	38.33	BS	8.3	66.6	6.6
S17	18:05, 13 Apr.	20:00, 14 Apr.	123.12	38.75	NYS	6.1	93.2	5.0
S18	14:31, 15 Apr.	14:33, 16 Apr.	123.08	38.15	NYS	7.1	79.8	4.7
S19	15:00, 16 Apr.	23:58, 16 Apr.	121.78	36.35	SYS	9.1	81.5	5.2

43 **Table S2.** Statistical summary of measured chemical components.

Component	SYS	NYS	BS
Amines (ng m^{-3})	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; μg m^{-3})	8.7 ± 4.4	13.0 ± 6.3	26.9 ± 4.9
Na^+	0.5 ± 0.5	0.7 ± 0.5	1.6 ± 0.3
NH_4^+	1.6 ± 0.7	2.4 ± 1.0	3.8 ± 0.5
K^+	0.2 ± 0.1	0.2 ± 0.1	0.5 ± 0.1
Mg^{2+}	0.0 ± 0.0	0.1 ± 0.1	0.3 ± 0.0
Ca^{2+}	0.3 ± 0.2	0.5 ± 0.2	1.9 ± 0.4
F^-	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
Cl^-	0.2 ± 0.4	0.6 ± 0.4	1.6 ± 0.6
NO_2^-	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
NO_3^-	1.8 ± 2.1	5.3 ± 2.7	10.6 ± 3.2
SO_4^{2-}	4.0 ± 2.2	3.4 ± 1.5	6.6 ± 0.9
Low molecular organic acids (μg m^{-3})	0.25 ± 0.12	0.20 ± 0.09	0.25 ± 0.02
CHO_2^-	0.03 ± 0.01	0.04 ± 0.03	0.05 ± 0.01
$C_2H_3O_2^-$	0.03 ± 0.01	0.03 ± 0.01	0.05 ± 0.02
$C_4H_4O_4^{2-}$	0.04 ± 0.04	0.04 ± 0.03	0.06 ± 0.01
$C_5H_6O_4^{2-}$	0.04 ± 0.03	0.05 ± 0.03	0.05 ± 0.00
$CH_3O_3S^-$ (MSA^-)	0.10 ± 0.08	0.05 ± 0.04	0.01 ± 0.02
Carbonaceous components (TC; μg m^{-3})	4.7 ± 1.5	5.9 ± 1.8	9.5 ± 1.8
Organic carbon (OC)	4.2 ± 1.4	5.1 ± 1.3	8.4 ± 1.6
Elemental carbon (EC)	0.4 ± 0.2	0.8 ± 0.4	1.0 ± 0.2
Organic compositions (ng m^{-3})	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} (≤ $C_{19:0}$)	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} (≤ C_{19alc})	1.4 ± 0.5	1.0 ± 0.6	1.5 ± 0.4

Component	SYS	NYS	BS
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

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Table S3. Concentrations of amines in marine 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	
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	
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	

Sea area	Sampling period	Particle size or gas	Concentrations of amines (ng m^{-3})						Reference
			MA	EA	DMA	IPA	PA	TMDEA	
Huaniao Island, China	November–December 2016	PM_{10}			15.1			8.4	(Zhou et al., 2019)
	March 2017				11.4			3.1	
	June–July 2017				32.2			27.5	
	August–September 2017				27.4			26.3	
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	
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	
Cape Verde, tropical Atlantic Ocean	November 2011 and 2013	Gas	0.8		4.5			< MDL	(Pinxteren et al., 2019)
		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)
Jeju Island, South Korea	March–April 2001	$PM_{2.5}$	13.5	3.1					(Yang et al., 2004)

Note: the SYS, NYS, BS, ECS and NWPO represent the South Yellow Sea, North Yellow Sea, Bohai Sea, East China Sea and Northwest Pacific Ocean, respectively.

47 **Table S4.** Correlation coefficients between amines and organic molecular tracers in
48 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

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

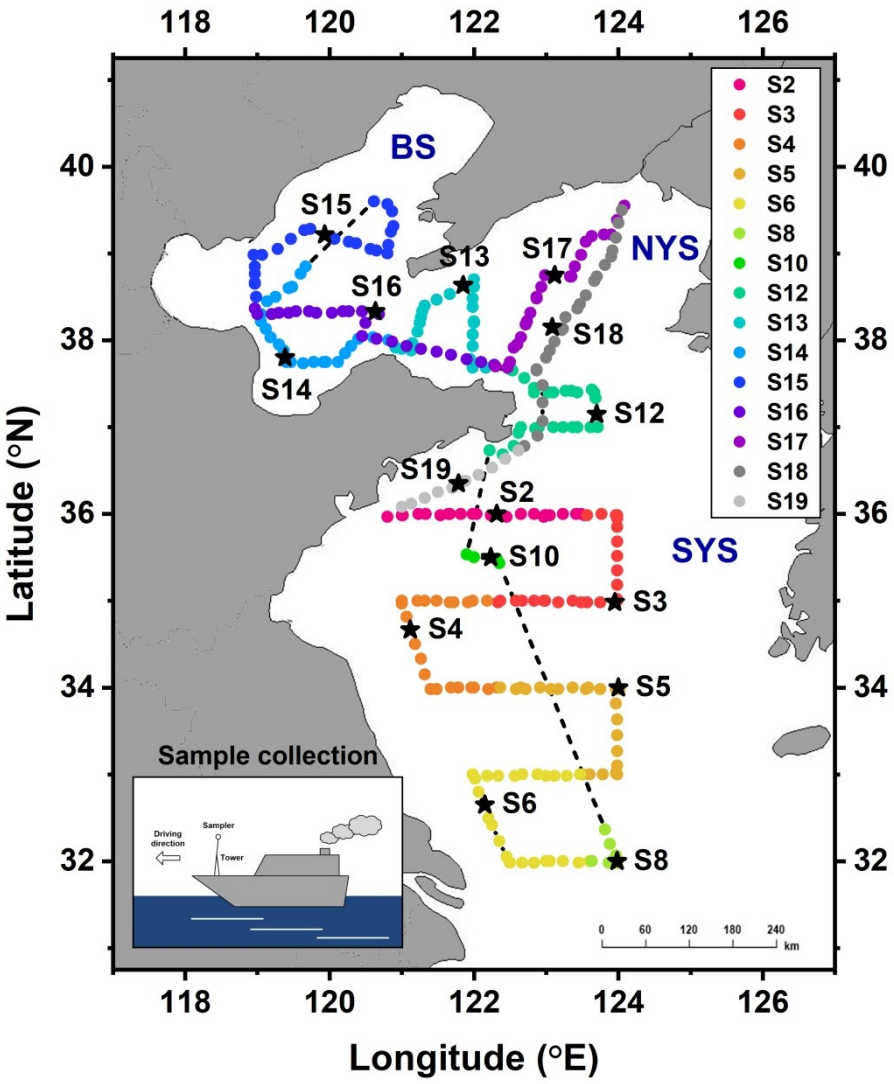


Figure S1. Map of the hourly sampling positions along the cruise track over the YS−BS, together with a schematic diagram of the sampling setup.

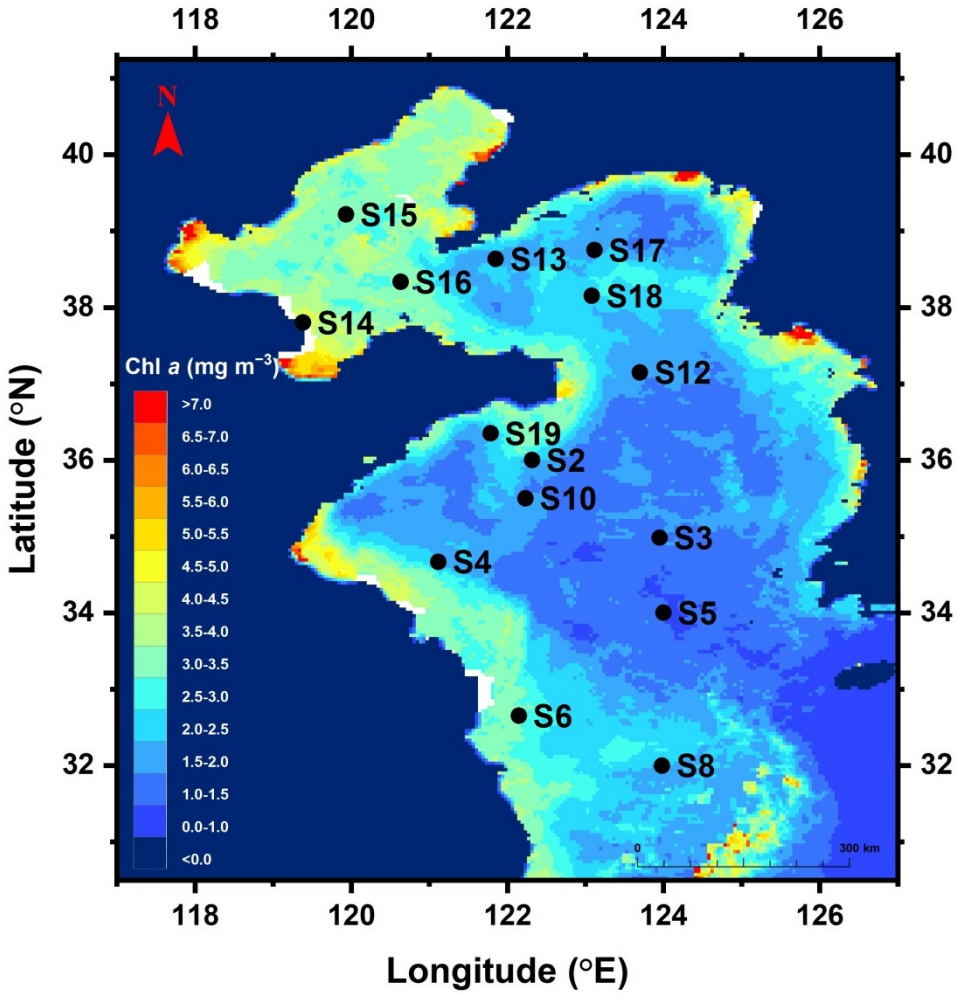
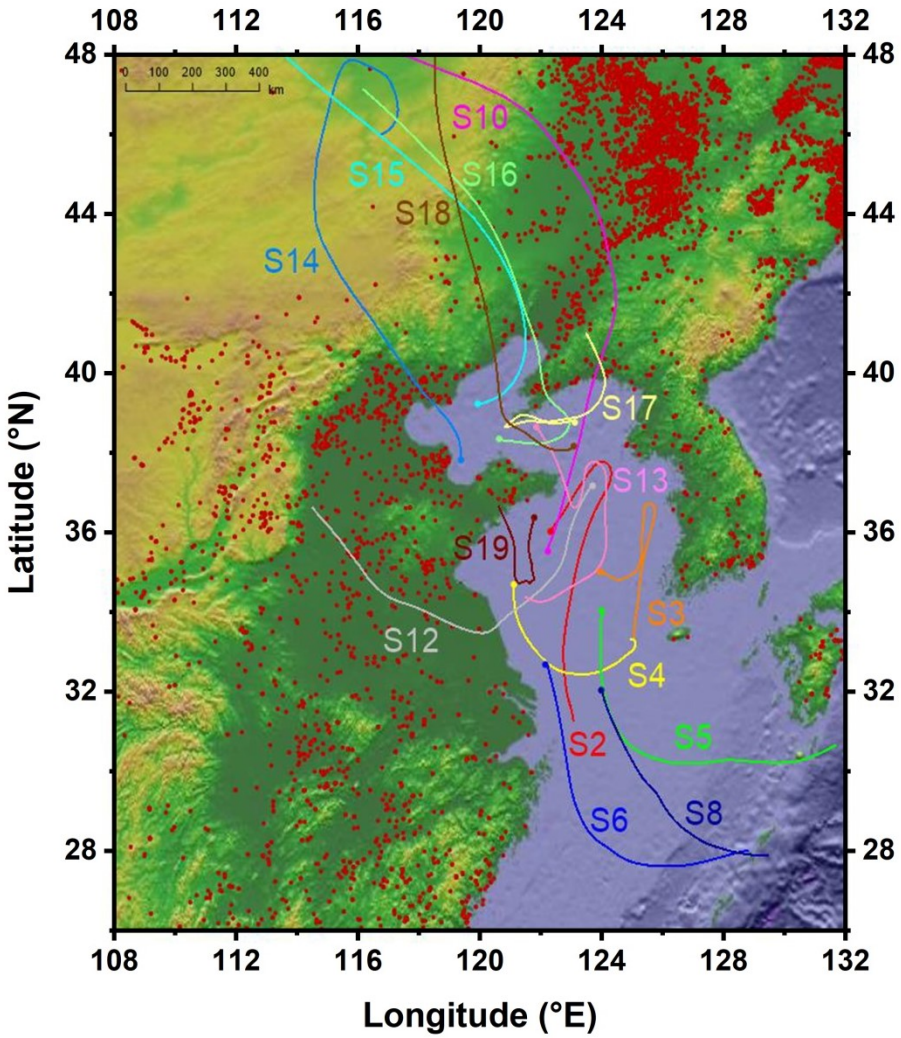


Figure S2. Map of chlorophyll *a* concentrations in surface seawater over the YS−BS during the cruise.



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61 **Figure S3.** 48 h backward air-mass trajectories starting from the midpoint of each
62 sampling period. Red dots indicate the fire spots during the cruise.

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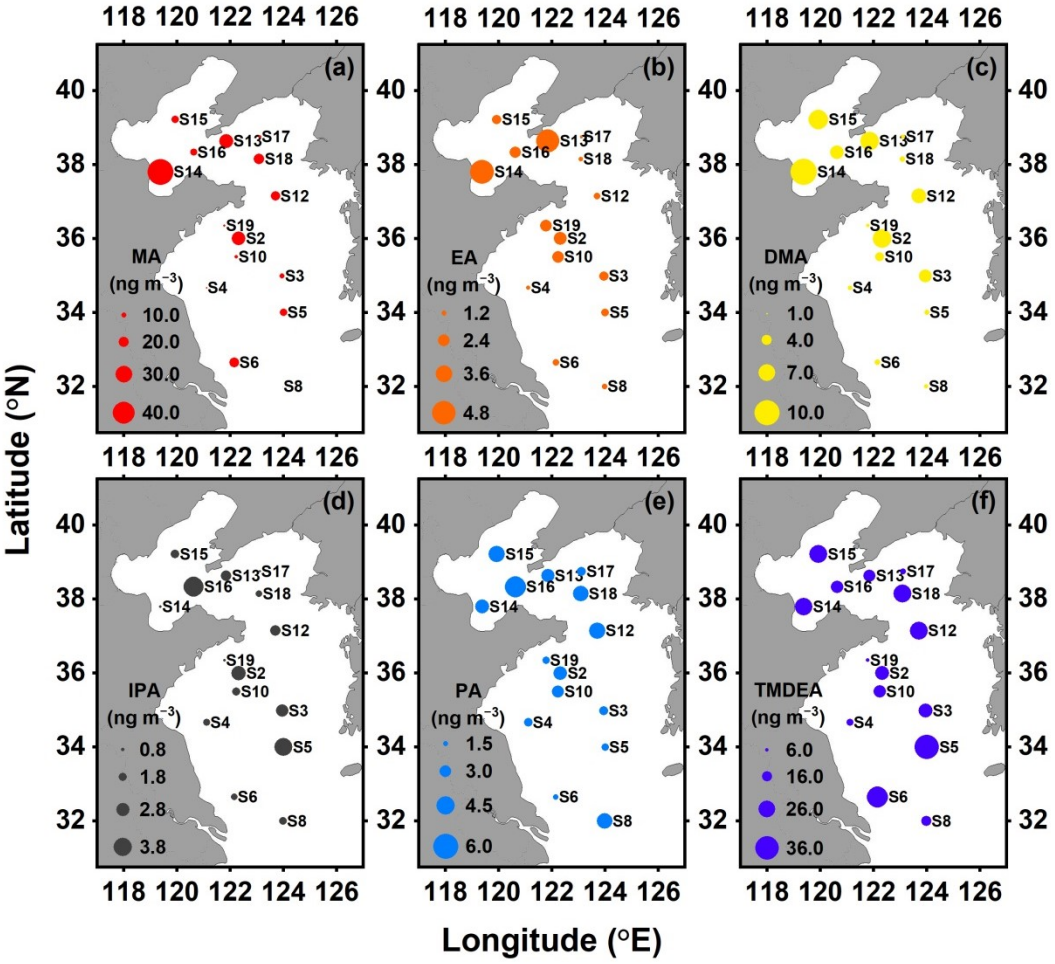

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65 **Figure S4.** Spatial distributions of MA (a), EA (b), DMA (c), IPA (d), PA (e), and
66 TMDEA (f) in TSP samples over the YS−BS.

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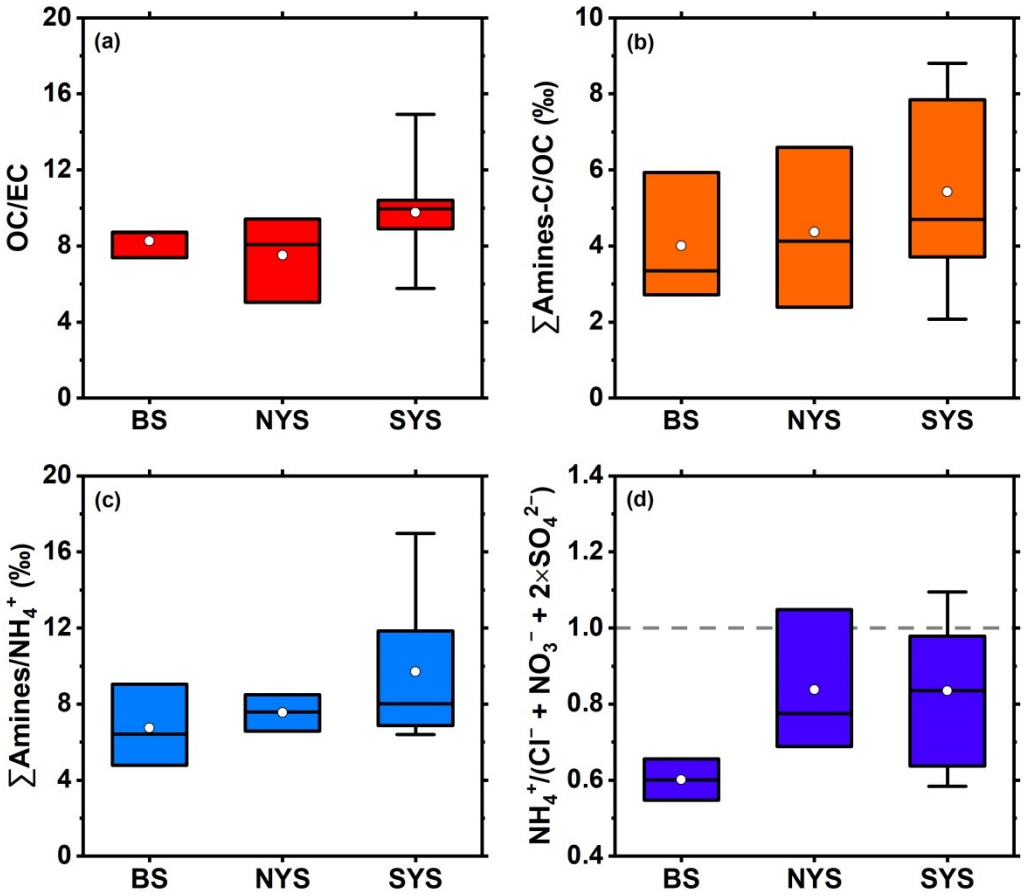
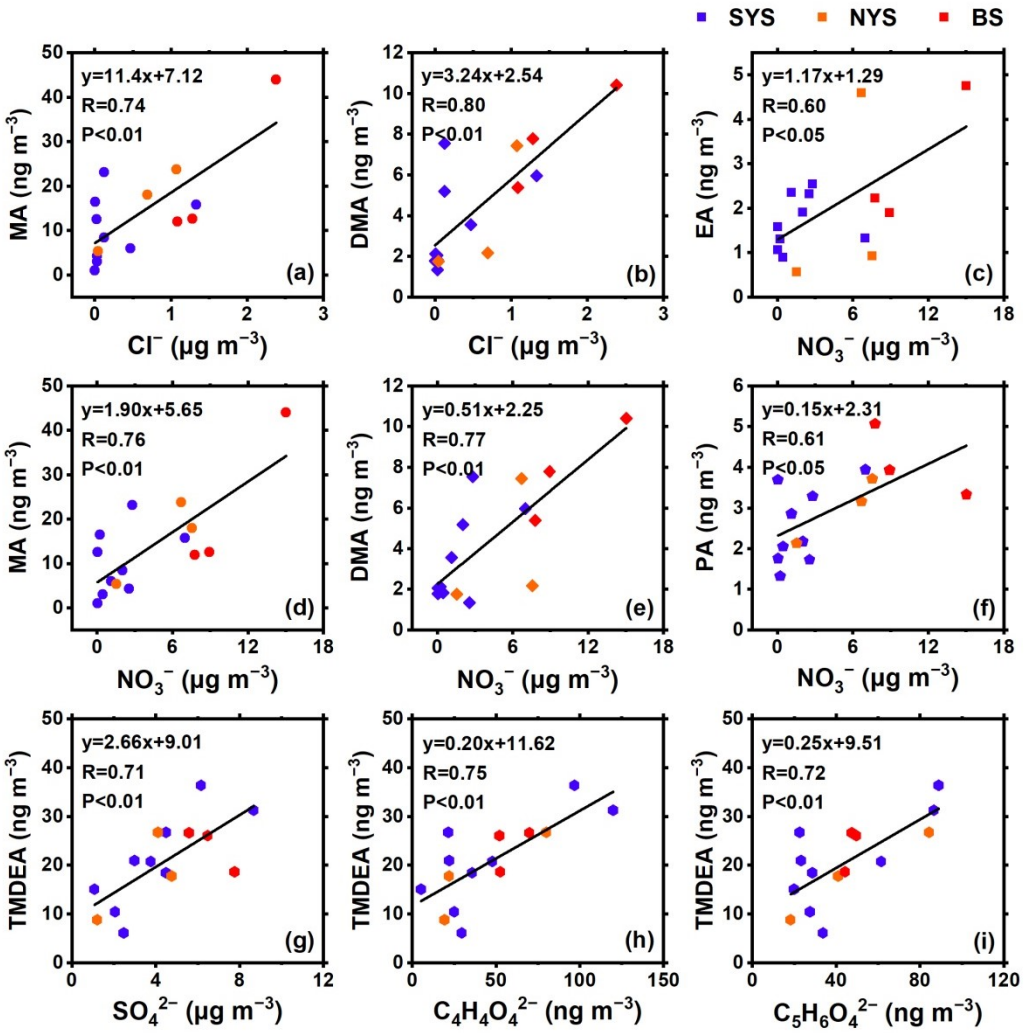


Figure S5. Variations of OC/EC ratios (a), ∑amines-C/OC ratios (b), ∑amines/NH_4^+ molar ratios (c), and $NH_4^+/(Cl^- + NO_3^- + 2\times SO_4^{2-})$ molar ratios (d) in TSP over the SYS, NYS, and BS. Lower and upper box boundaries indicate the 25th and 75th percentiles; lines within the box indicate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



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77 **Figure S6.** Linear regressions between amines and acidic species in TSP over the

78 SYS, NYS, and BS.

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