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Anthropogenic air pollutants strongly interact with natural aerosols over the eastern China seas: key processes, size distributions, and seasonalities

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Table S1. Mass concentrations of fine and coarse aerosols as well as associated chemical components over the eastern China seas in four cruises.

	Mass concentration (mean ± 1 standard deviation, unit: * μg m^{-3}, ** pg m^{-3}, others ng m^{-3})							
	Spring cruise		Summer cruise		Autumn cruise		Winter cruise	
	< 1.8 μm	> 1.8 μm	< 1.8 μm	> 1.8 μm	< 1.8 μm	> 1.8 μm	< 1.8 μm	> 1.8 μm
PM*	22.3 ± 12.8	17.9 ± 9.0	9.1 ± 4.0	11.6 ± 3.8	14.0 ± 8.9	26.6 ± 14.5	18.7 ± 7.5	12.9 ± 5.3
Na^{+}*	0.17 ± 0.08	1.31 ± 0.49	0.10 ± 0.06	1.36 ± 0.83	0.22 ± 0.14	3.42 ± 2.68	0.13 ± 0.05	1.23 ± 0.93
Cl^{-}*	0.25 ± 0.30	1.74 ± 0.71	0.10 ± 0.09	1.69 ± 1.25	0.24 ± 0.19	4.98 ± 4.58	0.15 ± 0.10	1.58 ± 1.45
SO_4^{2-}*	4.42 ± 1.17	0.74 ± 0.22	3.35 ± 1.25	0.87 ± 0.50	2.66 ± 1.42	0.99 ± 0.54	3.84 ± 1.33	0.52 ± 0.22
NO_3^{-}*	3.28 ± 4.51	2.25 ± 0.74	0.48 ± 0.73	1.73 ± 0.81	1.42 ± 2.51	3.05 ± 1.54	2.62 ± 2.24	1.64 ± 0.76
NH_4^{+}*	2.87 ± 2.07	0.31 ± 0.16	1.01 ± 0.69	0.27 ± 0.49	1.51 ± 1.29	0.25 ± 0.25	2.15 ± 1.98	0.19 ± 0.10
K^{+}	128 ± 92	72 ± 48	12 ± 10	36 ± 20	82 ± 66	109 ± 70	107 ± 62	39 ± 24
Mg^{2+}	71 ± 49	213 ± 76	12 ± 6	129 ± 93	61 ± 30	359 ± 225	43 ± 26	149 ± 64
Ca^{2+}	98 ± 110	246 ± 139	96 ± 51	137 ± 54	115 ± 57	419 ± 220	104 ± 65	184 ± 91
$C_2O_4^{2-}$	113 ± 42	14 ± 15	44 ± 42	25 ± 12	71 ± 40	26 ± 28	59 ± 31	12 ± 6
OC*	3.28 ± 1.91	0.70 ± 0.36	1.49 ± 0.85	0.17 ± 0.14				
Al	88 ± 58	424 ± 461	27 ± 33	56 ± 76	57 ± 69	495 ± 880	27 ± 13	99 ± 75
Ba	2.45 ± 1.88	7.75 ± 6.84	2.67 ± 2.32	2.59 ± 2.74	1.52 ± 1.31	6.54 ± 9.72	1.22 ± 0.60	2.04 ± 1.30
Ca	131 ± 114	517 ± 359	398 ± 458	436 ± 425	94 ± 57	667 ± 690	57 ± 35	227 ± 110
Cd	0.16 ± 0.21	0.06 ± 0.07	0.04 ± 0.07	0.03 ± 0.04	0.15 ± 0.10	0.07 ± 0.07	0.18 ± 0.14	0.05 ± 0.05
Ce	0.17 ± 0.16	0.77 ± 0.8	0.13 ± 0.16	0.18 ± 0.21	0.21 ± 0.20	1.44 ± 2.26	0.18 ± 0.10	0.34 ± 0.22
Co	0.13 ± 0.08	0.20 ± 0.17	0.06 ± 0.04	0.02 ± 0.01	0.07 ± 0.05	0.21 ± 0.33	0.07 ± 0.04	0.06 ± 0.03
Cu	5.76 ± 5.09	2.57 ± 2.01	1.15 ± 1.18	1.02 ± 1.18	1.73 ± 2.03	2.89 ± 4.45	1.68 ± 1.71	1.21 ± 1.03

	Mass concentration (mean ± 1 standard deviation, unit: * μg m^{-3}, ** pg m^{-3}, others ng m^{-3})							
	Spring cruise		Summer cruise		Autumn cruise		Winter cruise	
	< 1.8 μm	> 1.8 μm	< 1.8 μm	> 1.8 μm	< 1.8 μm	> 1.8 μm	< 1.8 μm	> 1.8 μm
Eu**	2.3 ± 1.7	9.7 ± 10.2	1.1 ± 1.5	1.1 ± 1.4	0.8 ± 0.7	5.1 ± 8.4	0.4 ± 0.2	1..0 ± 0.6
Fe	126 ± 114	424 ± 430	56 ± 61	83 ± 107	97 ± 103	412 ± 693	68 ± 50	129 ± 83
Ge	0.10 ± 0.08	0.02 ± 0.02	0.02 ± 0.02	0.01 ± 0.01	0.05 ± 0.04	0.02 ± 0.03	0.09 ± 0.06	0.01 ± 0.01
K	167 ± 97	175 ± 130	51 ± 65	113 ± 42	113 ± 80	265 ± 231	150 ± 73	104 ± 46
Mg	38 ± 22	308 ± 120	31 ± 22	214 ± 121	43 ± 35	541 ± 310	20 ± 19	173 ± 114
Mn	7.9 ± 6.5	8.5 ± 8.5	1.8 ± 2.3	1.5 ± 1.9	6.9 ± 7.7	9.9 ± 16.8	6.4 ± 5.0	3.4 ± 2.2
Mo	0.46 ± 0.35	0.20 ± 0.12	0.30 ± 0.13	0.16 ± 0.21	0.53 ± 0.97	0.26 ± 0.35	0.39 ± 0.34	0.34 ± 0.37
Na*	0.18 ± 0.06	1.65 ± 0.61	0.14 ± 0.02	1.60 ± 1.00	0.21 ± 0.11	3.43 ± 2.56	0.12 ± 0.06	1.24 ± 0.92
Ni	12.4 ± 15.5	1.9 ± 1.6	6.0 ± 3.6	1.2 ± 1.5	1.0 ± 0.7	1.1 ± 1.2	1.2 ± 1.0	0.6 ± 0.5
Pb	15.4 ± 12	3.7 ± 3.3	2.0 ± 3.4	1.1 ± 1.8	6.4 ± 3.9	2.1 ± 2.1	8.1 ± 5.2	1.5 ± 1.0
Rb	0.41 ± 0.29	0.48 ± 0.56	0.13 ± 0.13	0.23 ± 0.21	0.36 ± 0.29	0.81 ± 1.34	0.4 ± 0.27	0.17 ± 0.09
Sb	1.30 ± 1.42	0.23 ± 0.20	0.25 ± 0.40	0.22 ± 0.58	0.43 ± 0.45	0.11 ± 0.11	0.57 ± 0.41	0.12 ± 0.09
Se	1.49 ± 1.17	0.22 ± 0.13	0.80 ± 0.46	0.44 ± 0.20	0.85 ± 0.56	0.17 ± 0.11	0.81 ± 0.41	0.10 ± 0.07
Sr	0.69 ± 0.46	3.52 ± 2.03	0.49 ± 0.33	1.62 ± 0.64	0.55 ± 0.28	5.09 ± 3.62	0.39 ± 0.16	1.62 ± 0.80
V	13.1 ± 10.3	1.5 ± 1.5	12.8 ± 7	1.1 ± 0.5	0.7 ± 0.2	0.8 ± 1.3	0.6 ± 0.4	0.3 ± 0.2
Zn	42.9 ± 38.5	19.3 ± 16.9	31.7 ± 27.9	20.1 ± 18.0	22.1 ± 20.8	17.2 ± 24.4	18.9 ± 12.3	9.3 ± 6.1

Table S2. Statistical comparison between PMF-reconstructed and measured concentrations for each chemical species. The displayed metrices include the slope, intercept, and R^2 of the linear regression, and the normalized mean bias (NMB) between PMF-reconstructed and measured concentrations.

	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	$C_2O_4^{2-}$	Al	Ba	Ca	Cd	Ce	Co	Cu
Slope	0.982	0.801	0.717	0.897	0.732	0.998	0.731	0.746	0.740	0.743	0.940	0.700	0.703	0.808	0.836	0.614
Intercept	0.174	16.4	2.01	−1.08	4.28	−2.09	55.1	50.2	1.23	2.50	−0.0341	8.25	3.28E−3	7.50E−3	6.04E−4	0.0625
R^2	0.988	0.932	0.929	0.912	0.787	0.990	0.972	0.901	0.946	0.993	0.915	0.869	0.936	0.981	0.968	0.962
MNB	0.018	0.140	0.188	0.166	0.190	0.011	0.169	0.161	0.127	0.236	0.128	0.211	0.162	0.129	0.158	0.365

	Eu	Fe	Ge	K	Mg	Mn	Mo	Na	Ni	Pb	Rb	Sb	Se	Sr	V	Zn
Slope	0.702	0.893	0.709	0.715	0.817	0.798	0.534	0.977	0.519	0.760	0.703	0.636	0.716	1.009	0.762	0.629
Intercept	4.84E−5	1.54	1.20E−3	4.37	1.55	0.0989	0.0140	1.86	0.124	0.120	0.0124	0.0146	0.0219	−6.47E−3	0.0764	0.605
R^2	0.959	0.977	0.893	0.941	0.983	0.970	0.961	0.981	0.979	0.939	0.980	0.964	0.949	0.986	0.975	0.960
NMB	0.270	0.066	0.193	0.169	0.166	0.140	0.454	0.014	0.397	0.138	0.172	0.234	0.102	0.011	0.119	0.320

Table S3. All-season average fractional contributions (%) of each PMF factor to the total mass concentration of each species.

	Dust	Primary anthropogenic emissions	Continental secondary + burning	Aged continental and marine secondary	Shipping emission	Fresh sea salt	Aged sea salt
Na^{+}	0	0.93	0.32	1.26	0.47	70.33	26.68
NH_4^{+}	0.60	0	56.53	38.39	4.48	0	0
K^{+}	1.97	11.74	36.17	10.49	0	31.17	8.46
Mg^{2+}	8.90	0	4.81	1.62	0.59	57.06	27.03
Ca^{2+}	38.57	7.35	2.66	6.11	3.03	33.58	8.69
Cl^{-}	2.54	0	1.81	0	1.01	94.64	0
NO_3^{-}	12.69	0	44.15	0	0	0	43.17
SO_4^{2-}	0	0.28	29.48	48.45	9.09	7.89	4.81
$C_2O_4^{2-}$	3.68	0	0	88.13	0	0	8.19
Al	88.83	3.05	1.45	1.34	0.25	1.59	3.49
Ba	77.17	10.51	2.67	0.89	4.09	1.56	3.12
Ca	61.12	0	0.10	7.47	5.32	17.12	8.88
Cd	5.83	72.50	17.43	2.53	1.48	0.23	0
Ce	82.39	8.39	1.60	0.46	0.98	1.79	4.39
Co	62.12	9.21	9.07	5.53	9.40	1.84	2.84
Cu	37.38	29.96	14.75	6.66	7.59	2.02	1.63
Eu	82.56	5.04	1.49	3.12	3.55	2.22	2.02
Fe	76.53	12.20	5.02	0.69	1.55	1.11	2.91
Ge	11.82	7.39	45.45	33.12	0.91	0.50	0.80
K	20.36	25.31	16.21	3.01	0.86	19.62	14.64
Mg	21.55	0.82	0	1.54	0.61	50.73	24.76
Mn	51.81	31.96	12.62	0	1.00	0.83	1.79
Mo	16.42	41.99	16.55	5.86	15.17	1.41	2.60

	Dust	Primary anthropogenic emissions	Continental secondary + burning	Aged continental and marine secondary	Shipping emission	Fresh sea salt	Aged sea salt
Na	1.56	1.14	0	1.13	0.52	65.03	30.63
Ni	15.94	12.75	3.32	5.96	60.10	1.67	0.24
Pb	6.35	52.93	33.09	6.84	0.72	0	0.07
Rb	42.51	25.10	16.40	7.74	0	4.23	4.02
Sb	5.23	38.43	36.13	13.71	4.46	0.87	1.17
Se	0	18.48	20.96	40.05	8.40	0	12.10
Sr	45.22	2.67	1.25	1.66	1.55	28.77	18.88
V	8.57	3.84	0.34	0	84.81	0	2.43
Zn	17.12	51.49	15.38	0	10.10	0	5.91

Table S4. All-season average fractional contributions (%) of each PMF factor to the fine-mode (< 1.8 μm) mass concentration of each species.

	Dust	Primary anthropogenic emissions	Continental secondary + burning	Aged continental and marine secondary	Shipping emission	Fresh sea salt	Aged sea salt
Na^+	0	9.56	3.41	15.39	6.63	34.76	30.25
NH_4^+	0.08	0	52.97	41.43	5.53	0	0
K^+	0.43	18.24	58.12	19.41	0	2.34	1.45
Mg^{2+}	8.59	0	33.82	13.09	5.43	18.72	20.35
Ca^{2+}	18.51	24.85	9.31	24.64	13.97	5.48	3.25
Cl^-	4.40	0	22.84	0	16.92	55.83	0
NO_3^-	3.44	0	87.41	0	0	0	9.14
SO_4^{2-}	0	0.27	29.96	56.71	12.17	0.37	0.52
$C_2O_4^{2-}$	0.49	0	0	98.66	0	0	0.85
Al	64.45	15.59	7.65	8.18	1.75	0.39	1.98
Ba	35.01	33.60	8.84	3.39	17.82	0.24	1.10
Ca	32.44	0	0.37	33.30	27.13	3.09	3.68
Cd	0.86	75.20	18.70	3.13	2.09	0.01	0
Ce	48.33	34.67	6.85	2.27	5.51	0.36	2.01
Co	18.67	19.50	19.88	13.95	27.14	0.19	0.67
Cu	7.68	43.36	22.09	11.49	14.98	0.14	0.26
Eu	43.09	18.55	5.67	13.67	17.80	0.39	0.82
Fe	34.42	38.66	16.45	2.59	6.70	0.17	1.02
Ge	1.73	7.63	48.52	40.72	1.28	0.02	0.09
K	5.52	48.39	32.07	6.85	2.24	1.81	3.10
Mg	26.07	6.98	0	15.68	7.04	20.87	23.37
Mn	13.62	59.22	24.19	0	2.52	0.07	0.37
Mo	2.60	46.93	19.14	7.80	23.13	0.08	0.32

	Dust	Primary anthropogenic emissions	Continental secondary + burning	Aged continental and marine secondary	Shipping emission	Fresh sea salt	Aged sea salt
Na	2.23	11.46	0	13.49	7.16	31.56	34.10
Ni	2.10	11.85	3.20	6.60	76.15	0.07	0.02
Pb	0.93	54.45	35.22	8.38	1.01	0	0.01
Rb	10.41	43.29	29.28	15.90	0	0.35	0.77
Sb	0.75	38.76	37.71	16.48	6.13	0.04	0.13
Se	0	18.35	21.54	47.40	11.38	0	1.33
Sr	35.75	14.87	7.19	11.01	11.80	7.73	11.64
V	1.00	3.17	0.29	0	95.32	0	0.22
Zn	2.88	61.11	18.89	0	16.34	0	0.78

Table S5. All-season average fractional contributions (%) of each PMF factor to the coarse-mode (> 1.8 μm) mass concentration of each species.

	Dust	Primary anthropogenic emissions	Continental secondary + burning	Aged continental and marine secondary	Shipping emission	Fresh sea salt	Aged sea salt
Na^+	0	0.20	0.03	0.15	0.03	73.60	25.99
NH_4^+	5.09	0	51.13	41.12	2.66	0	0
K^+	3.71	4.92	7.21	2.48	0	65.21	16.48
Mg^{2+}	8.82	0	0.50	0.20	0.04	62.76	27.68
Ca^{2+}	43.96	1.86	0.32	0.87	0.24	42.51	10.24
Cl^-	2.35	0	0.18	0	0.07	97.40	0
NO_3^-	20.46	0	7.54	0	0	0	72.00
SO_4^{2-}	0	0.26	13.21	25.71	2.68	37.09	21.06
$C_2O_4^{2-}$	15.86	0	0	47.63	0	0	36.51
Al	93.28	0.71	0.16	0.18	0.02	1.86	3.79
Ba	90.64	2.74	0.33	0.13	0.33	2.03	3.79
Ca	67.43	0	0.01	1.03	0.41	20.98	10.13
Cd	23.83	65.88	7.54	1.30	0.42	1.04	0
Ce	90.45	2.05	0.19	0.06	0.07	2.19	4.99
Co	86.93	2.87	1.34	0.97	0.92	2.86	4.11
Cu	73.44	13.09	3.07	1.64	1.04	4.42	3.31
Eu	92.73	1.26	0.18	0.44	0.28	2.77	2.34
Fe	90.88	3.22	0.63	0.10	0.13	1.46	3.58
Ge	49.50	6.89	20.15	17.39	0.27	2.34	3.46
K	31.29	8.65	2.64	0.58	0.09	33.48	23.27
Mg	20.74	0.18	0	0.19	0.04	54.22	24.64
Mn	82.12	11.27	2.12	0	0.11	1.46	2.93
Mo	48.93	27.82	5.22	2.19	3.15	4.68	8.02

	Dust	Primary anthropogenic emissions	Continental secondary + burning	Aged continental and marine secondary	Shipping emission	Fresh sea salt	Aged sea salt
Na	1.47	0.24	0	0.13	0.03	68.21	29.91
Ni	60.93	10.84	1.34	2.86	15.99	7.08	0.96
Pb	28.09	52.07	15.49	3.79	0.22	0	0.33
Rb	71.26	9.35	2.91	1.63	0	7.88	6.97
Sb	23.99	39.18	17.54	7.88	1.42	4.45	5.54
Se	0	16.81	9.07	20.53	2.39	0	51.19
Sr	46.27	0.61	0.14	0.21	0.11	32.69	19.97
V	47.92	4.78	0.20	0	33.03	0	14.06
Zn	46.26	30.93	4.40	0	1.90	0	16.51

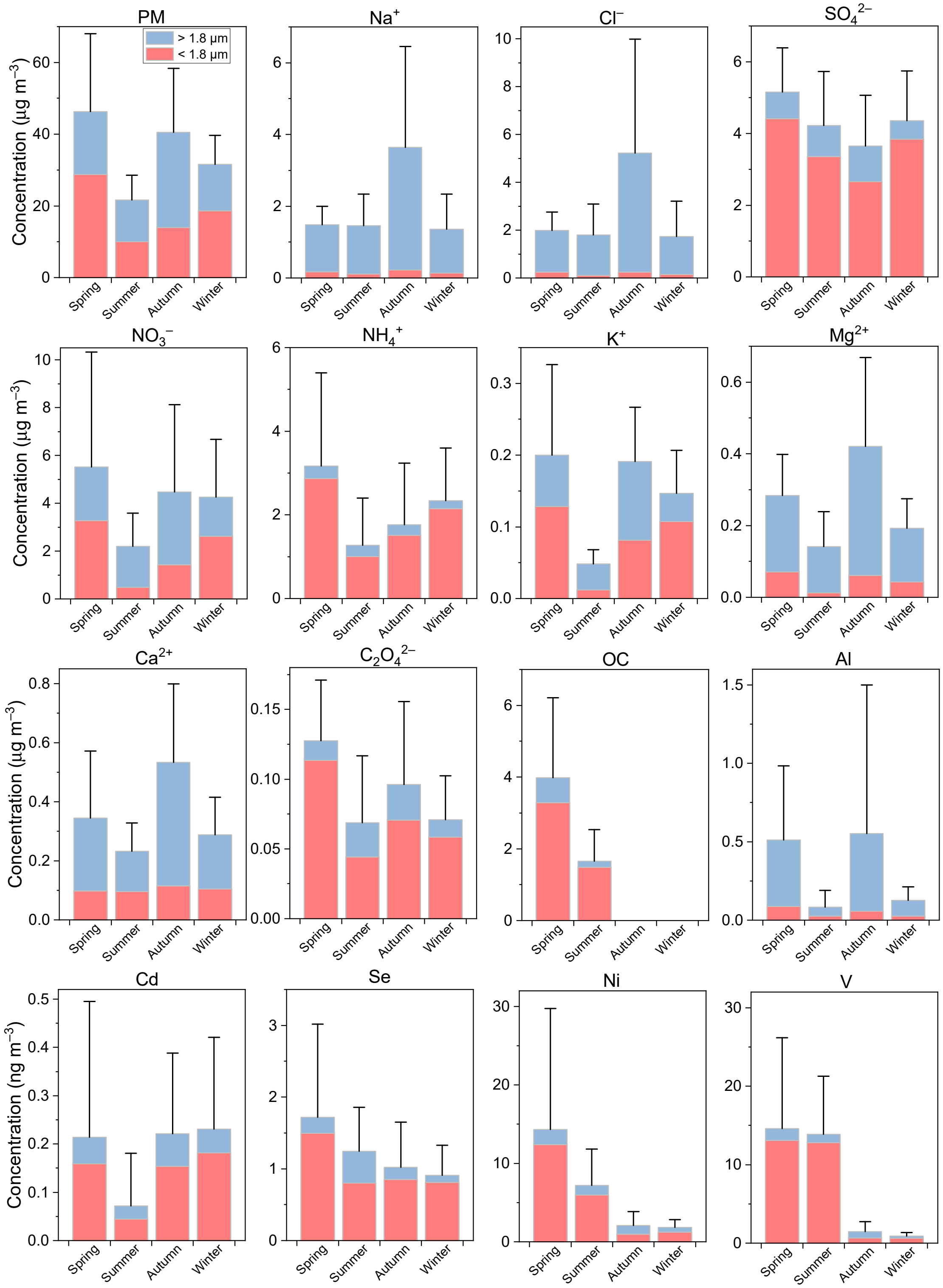

Figure S1. Mass concentrations of fine and coarse aerosols as well as typical chemical components in four cruises. Error bars represent the standard deviation of total concentrations across all samples collected during each cruise.

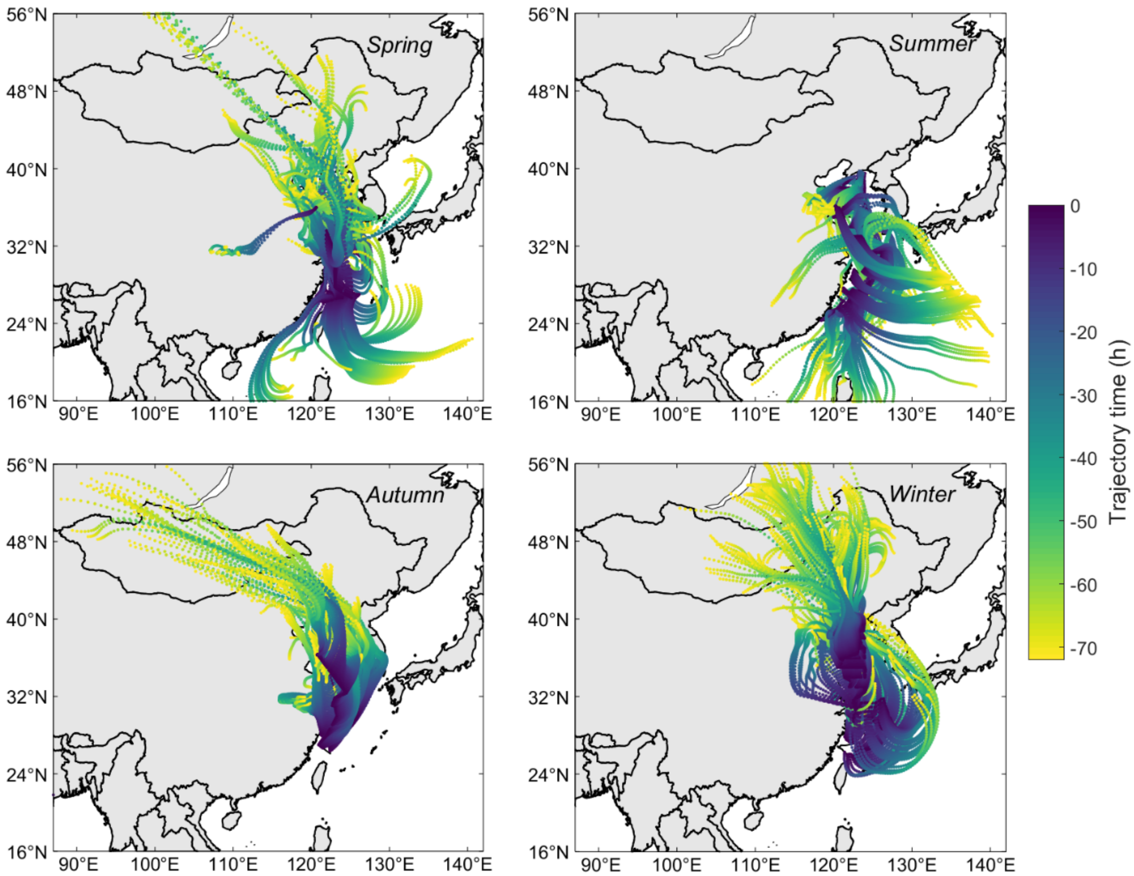

Figure S2. 72-hour air mass backward trajectories arriving at the sampling sites during the four cruise campaigns. The color of each trajectory point indicates the time before arrival. Only trajectories with arrival times falling within the MOUDI sampling periods are shown.

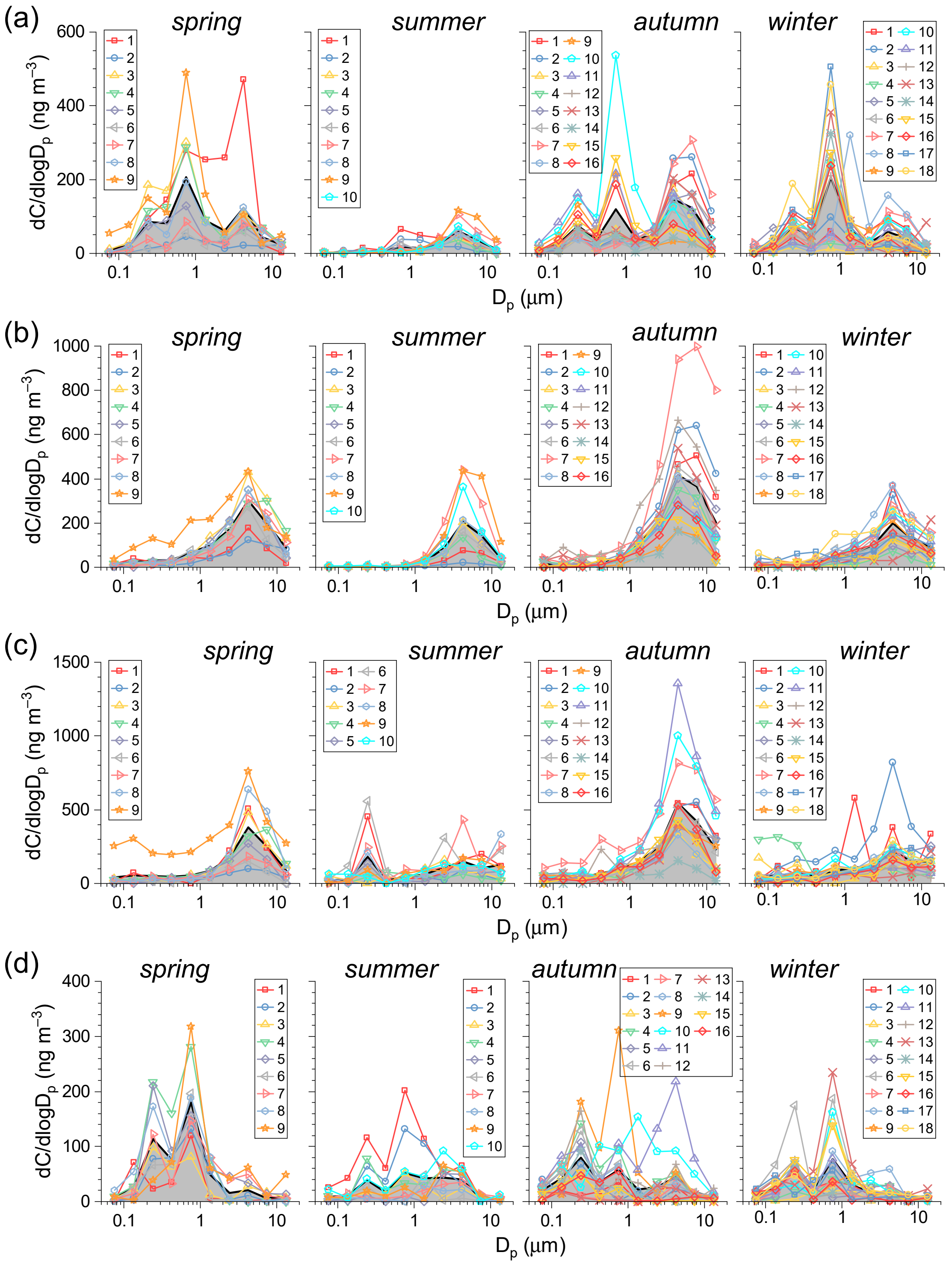

Figure S3. Size distributions of (a) K^+, (b) Mg^{2+}, (c) Ca^{2+}, and (d) $C_2O_4^{2-}$ during four cruises. Each number represents a sample set. The thick black line with gray area below represents the average size distribution for each cruise.

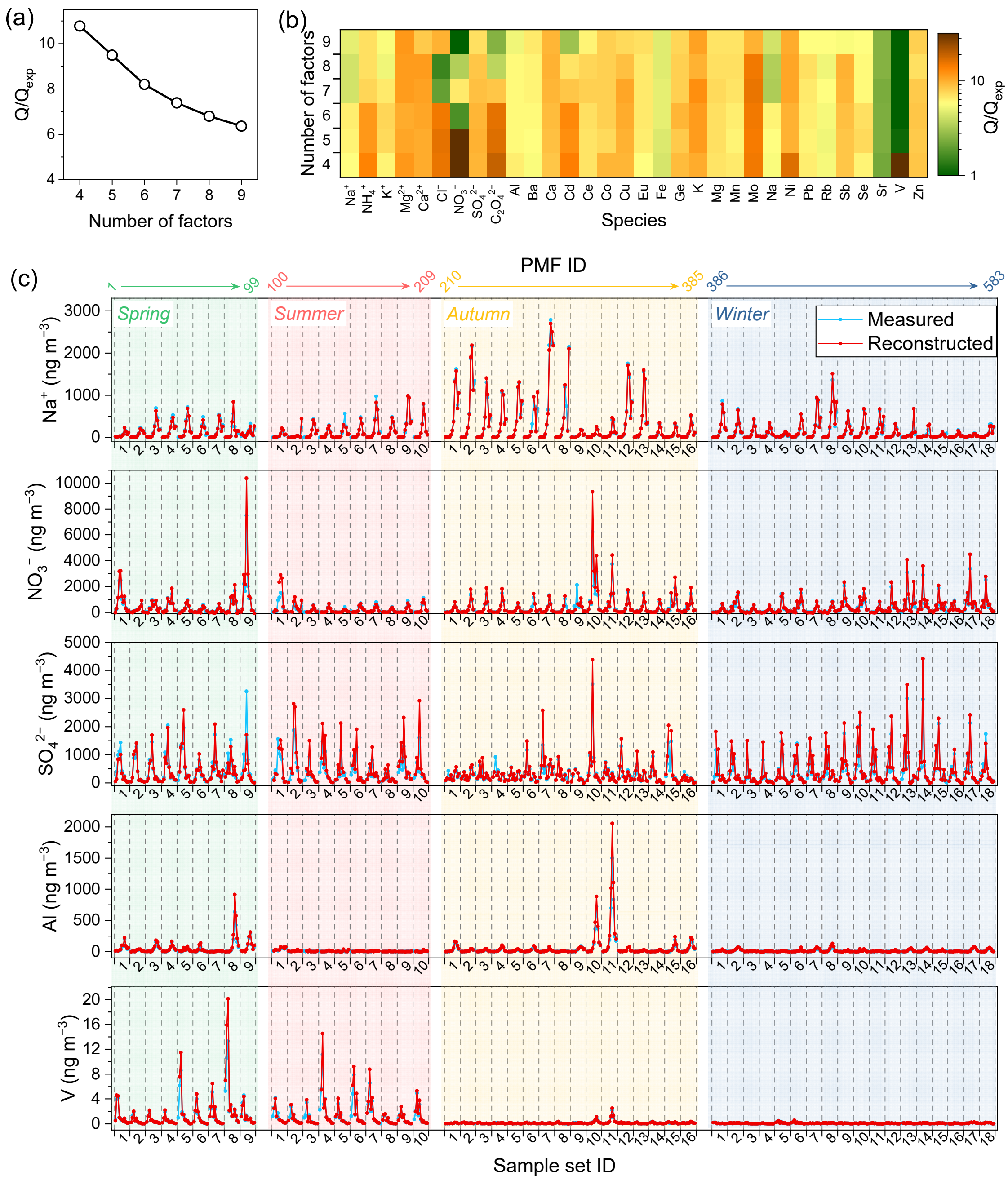

Figure S4. (a) Trend of the Q/Q_{exp} value for the entire dataset as a function of the number of PMF factors. (b) Same as panel (a) but for each chemical species. (c) Time series of measured and PMF-reconstructed concentrations of five representative species. Each dot represents the concentration within a specific size bin of a sample set. Statistical comparisons between measured and PMF-reconstructed concentrations for other species are provided in Table S2.

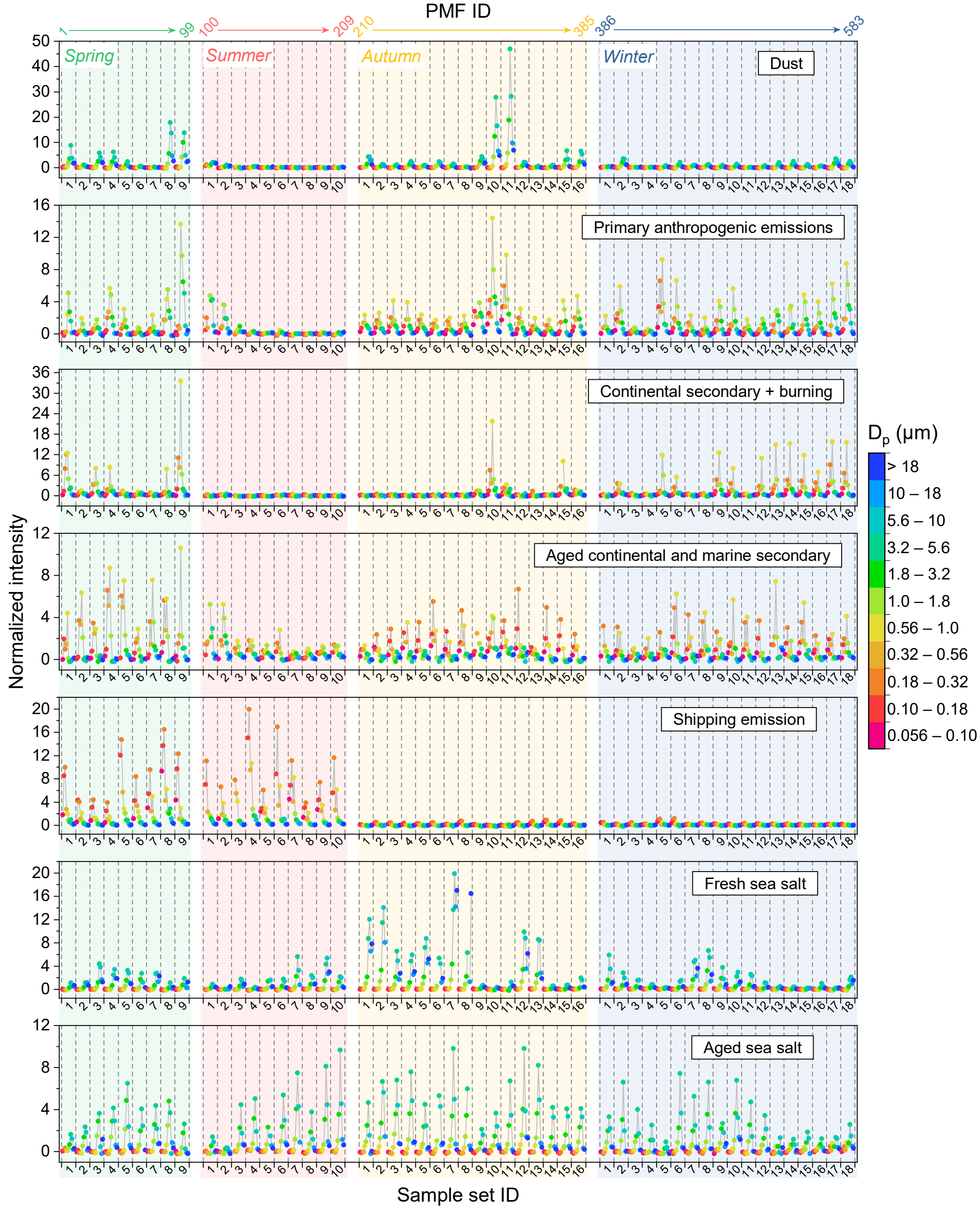

Figure S5. Raw series of the normalized contribution intensity of different PMF factors for each size range in each sample set.

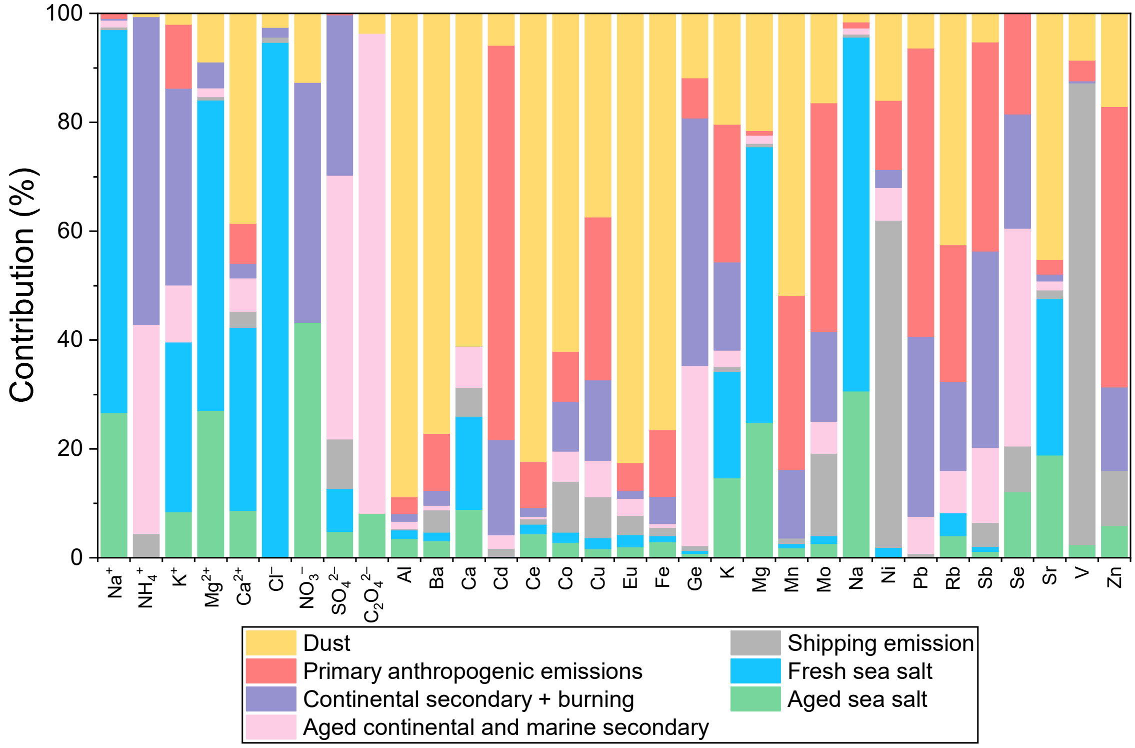

Figure S6. The relative contributions of different sources to the size-integrated concentration of each aerosol chemical species across all four seasons.

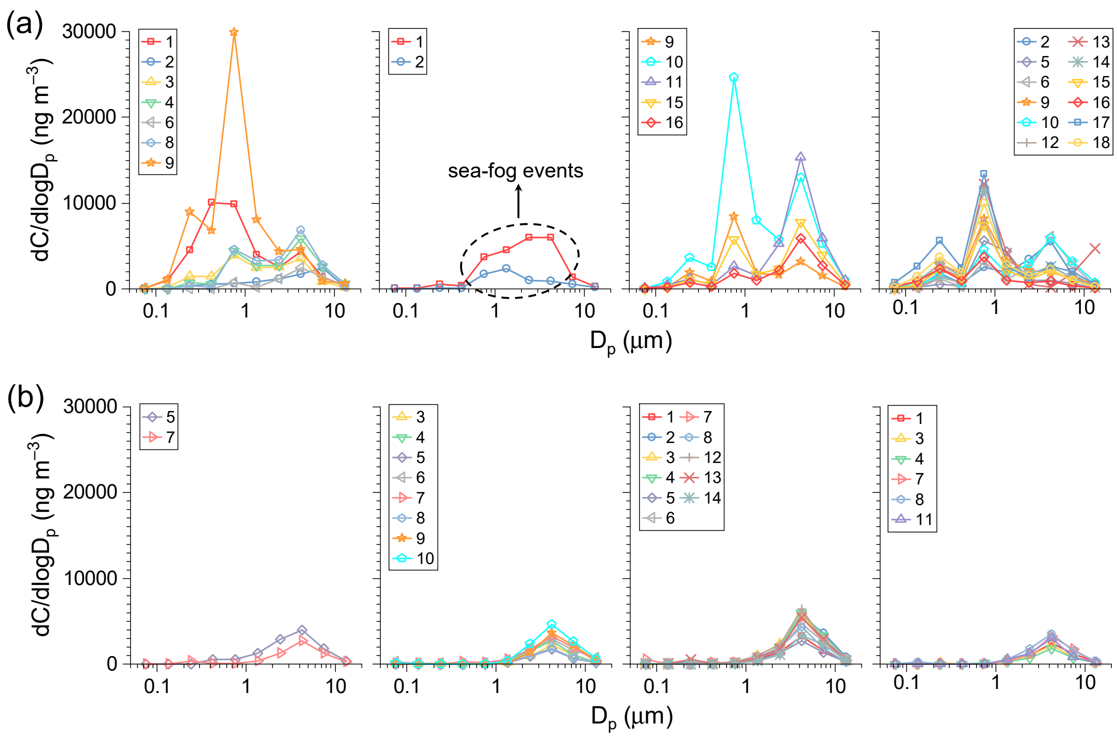

Figure S7. Two distinct types of NO_3^- size distribution: (a) bimodal or trimodal distribution, indicating a strong influence from continental anthropogenic pollution, with relatively fresh pollution aerosols; (b) unimodal distribution, suggesting a relatively weak direct influence from continental pollution, with highly aged pollution aerosols.

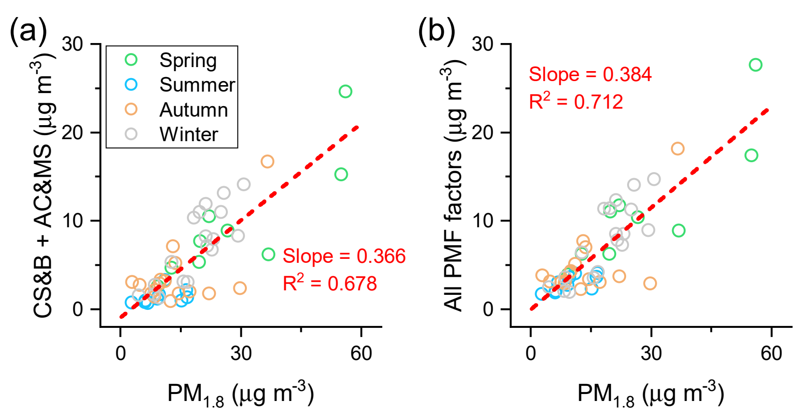

Figure S8. Correlations between $PM_{1.8}$ mass concentrations and the total concentrations of PMF species in sub-1.8 μm particles contributed by (a) CS&B and AC&MS, and (b) all PMF factors. Here PMF species correspond to all chemical species included in PMF analysis but excluding duplicate species (e.g., Na^+ is excluded because elemental Na is also included). Different colors represent different seasons.

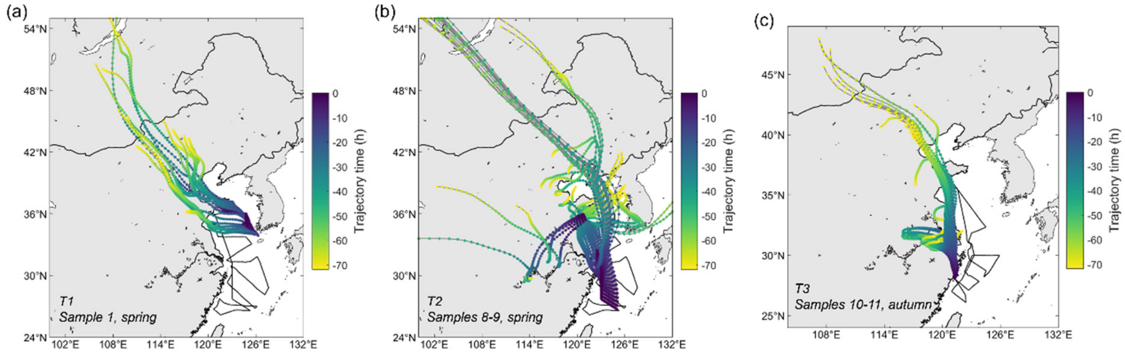

Figure S9. 72-hour air mass backward trajectories during three co-transport events of dust and continental pollution aerosols: (a) Sample 1 from the spring campaign, (b) Samples 8-9 from the spring campaign, and (c) Samples 10-11 from the autumn campaign. The color of each trajectory point indicates the time before arrival at the sampling sites.

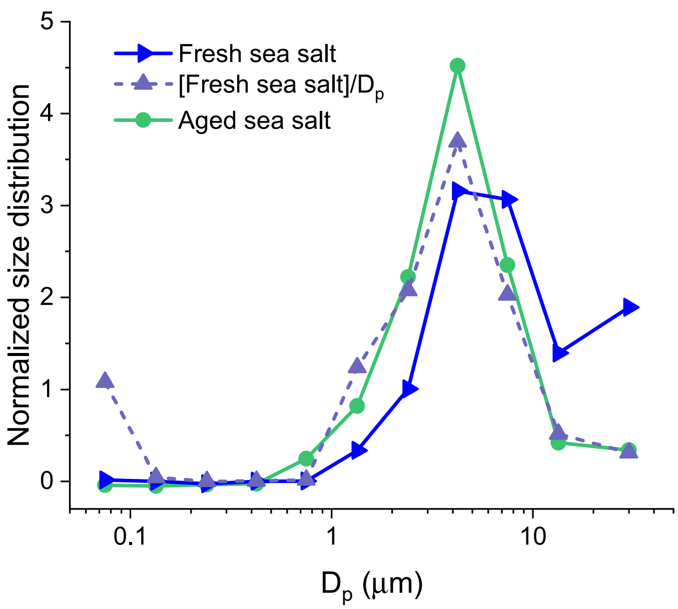

Figure S10. Normalized size distributions of average concentrations from fresh sea salt and aged sea salt PMF factors, along with the ratio of fresh sea salt to particle diameter ([Fresh sea salt]/D_p), which serves as a proxy for the surface area of fresh sea salt. The abnormally high value of [Fresh sea salt]/D_p in the smallest size bin is likely due to large measurement and PMF uncertainties associated with the extremely low sea salt concentrations in that range.

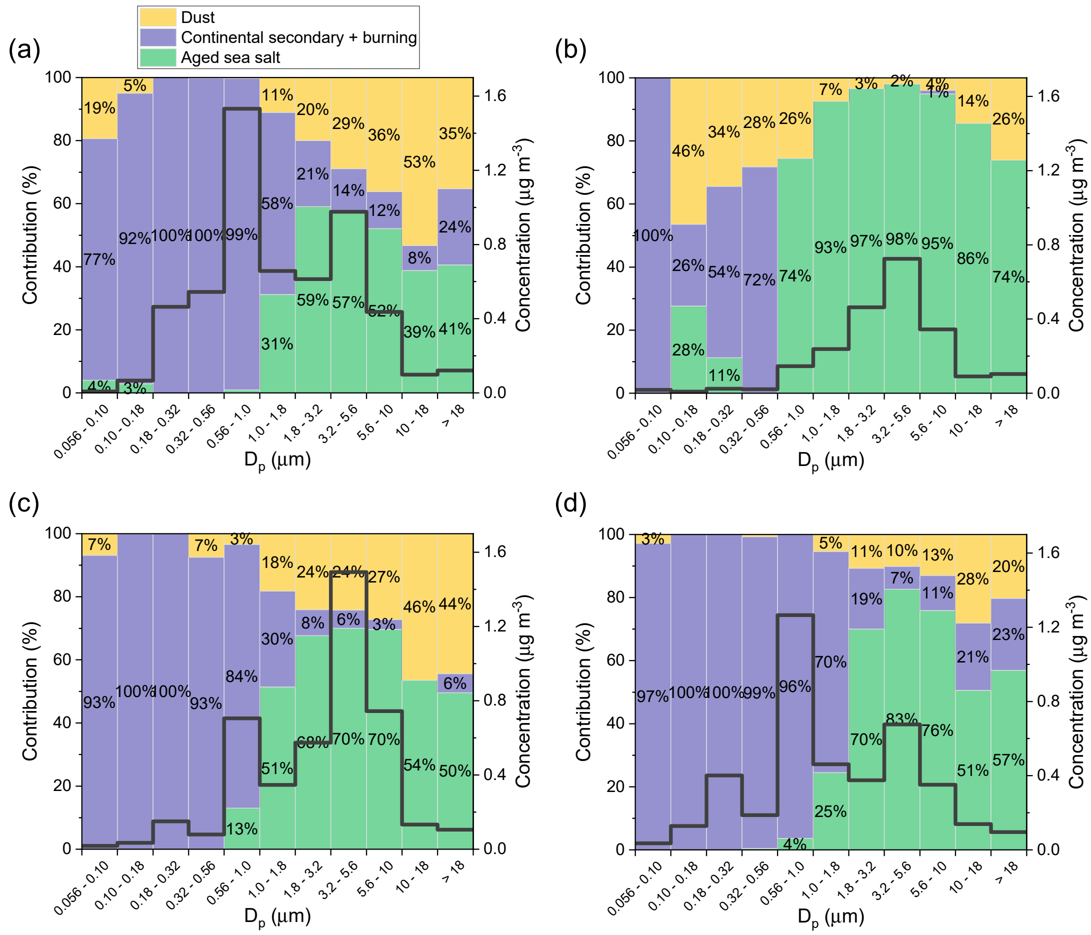

Figure S11. Fractional contributions of different sources to NO_3^- in size-segregated aerosols during (a) spring, (b) summer, (c) autumn, and (d) winter cruises. Thick black lines represent the average measured NO_3^- concentrations in each size bin. Note that source contributions in size bins with very low NO_3^- concentrations (e.g., D_p < 0.56 μm in summer) are subject to considerable uncertainty and may not be statistically robust; however, this has minimal impact on the overall source profile.

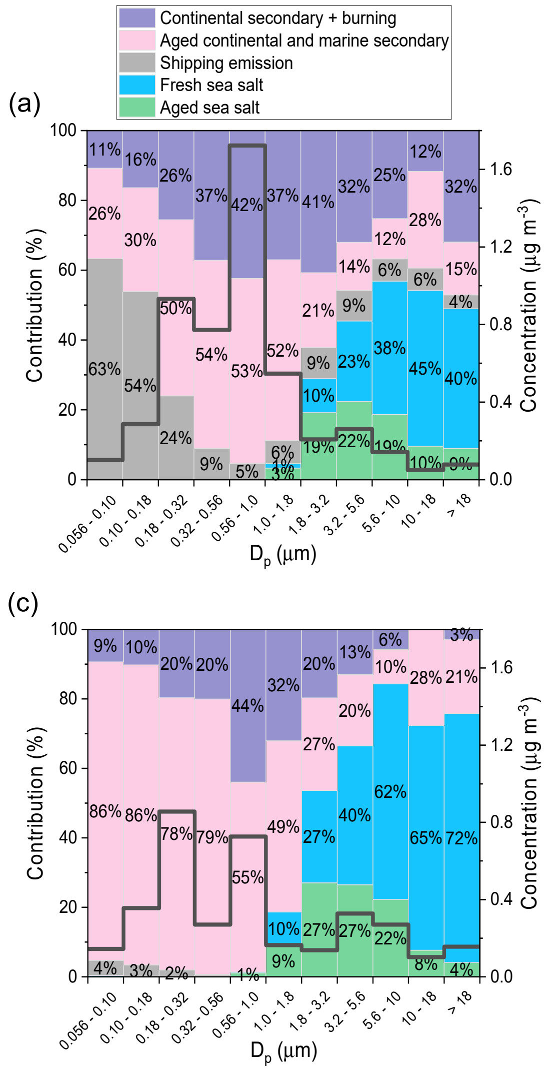

Figure S12. Same as Fig. S11 but for SO_4^{2-}.