



Supplement of

The concentration, source and deposition flux of ammonium and nitrate in atmospheric particles during dust events at a coastal site in northern China

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Table S1. The concentrations and the increase ratio^a of TSP, Al, Fe, nss-Ca, NH₄⁺ and NO₃⁻ in the aerosols samples collected at the Baguanshan site in the coastal region of the Yellow Sea.

Sampling Month	Sample category	Sampling number	TSP		Al		Fe		nss-Ca		NH ₄ ⁺		NO ₃ ⁻	
			Concentration n (μg/m ³)	Increase ratio (%)	Concentration on (μg/m ³)	Increase ratio (%)	Concentration on (μg/m ³)	Increase ratio (%)	Concentration on (μg/m ³)	Increase ratio (%)	Concentration on (μg/m ³)	Increase ratio (%)	Concentration on (μg/m ³)	Increase ratio (%)
Mar., 2008	Dust days (DD)	20080301	526.7	134	21.3	136	14.1	117	25.2	76	12.7	51	20.5	63
	Reference sample (RS)	20080315	409.5	82	17.5	94	10.3	58	14.6	2	13.9	65	19.5	55
	Reference sample (RS)	20080316	225.1		9.0		6.5		14.3		8.4		12.6	
Apr., 2008	Dust days (DD)	20080425	622.2	353	33.2	403	10.6	126	63.1	507	2.0	-72	6.8	-69
	Reference sample (RS)	20080424	137.5		6.6		4.7		10.4		7.2		21.7	
May, 2008	Dust days (DD)	20080528	2578.7	1151	182.8	1977	96.1	4078	79.2	350	2.7	-84	9.2	-66
	Reference sample (RS)	20080529	2313.8	1023	132.7	1408	70.5	2965	46.8	166	4.8	-71	17.5	-36
	Reference sample (RS)	20080522	206.1		8.8		2.3		17.6		16.6		27.4	
Mar., 2009	Dust days (DD)	20090316	688.4	630	24.2	1244	14.8	679	29.6	323	17.2	473	15.9	835
	Reference sample (RS)	20090306	94.3		1.8		1.9		7.0		3.0		1.7	
Mar., 2010	Dust days (DD)	20100315	501.1	82	25.6	173	14.8	95	15.2	-13	4.3	79	5.4	-25
	Reference sample (RS)	20100320	3856.7	1303	205.4	2090	116.3	1430	151.1	768	3.4	42	5.5	-24
	Reference sample (RS)	20100321	518.6	89	25.8	175	15.3	101	19.2	10	9.4	292	16.5	129
	Reference sample (RS)	20100324	274.8		9.38		7.6		17.4		2.4		7.2	

	Dust days (DD)	20110319	938.6	384	39.3	602	26.3	498	21.2	71	9.4	-28	12.3	-5
Mar., 2011	Reference sample (RS)	20110308	194.1		5.6		4.4		12.4		13.1		13.0	
Apr., 2011	Dust days (DD)	20110418	557.9	121	26.6	144	22.3	260	14.2	51	6.6	22	3.8	-32
	Reference sample (RS)	20110416	252.3		10.9		6.2		9.4		5.4		5.6	
May, 2011	Dust days (DD)	20110501	501.8	124	27.0	68	16.2	184	12.1	329	5.3	-48	4.5	-70
	Reference sample (RS)	20110502	809.7	261	48.1	199	27.5	382	21.3	656	11.0	8	21.0	38
Mean of all samples	Dust days (DD)	1140.3	403	62.3	615	35.1	777	39.4	228	7.9			12.1	
	Reference sample (RS)	201.1		8.5		4.9		11.4		8.3			13.1	

^aIncrease in percentage is calculated by the formula $(C_{DDi} - C_{RSi})/C_{RSi} \times 100\%$, where C_{DDi} and C_{RSi} was the concentration of substance i in each dust day sample and reference sample, respectively, for those samples where the increase in percentage is positive.

Table S2. The Spearman correlation coefficients of NO_3^- -N and NH_4^+ -N with average transport speed, transport distance over the sea, transport altitude, transport time in polluted region, air temperature, RH and NO_2 concentration during the transport of dust days samples (n=13).

	S ^a	Distance over sea	AMLD ^b	Transport altitude	Time in polluted region ^c	T ^d	RH	NO_2
NO ₃ ⁻ -N Correlation Coefficient	.154	-.317	.148	.088	.619*	-.044	-.646*	.352
	.616	.292	.629	.775	.024	.887	.017	.239
NH ₄ ⁺ -N Correlation Coefficient	.154	-.137	-.379	.247	.495	-.297	-.322	.159
	.616	.654	.201	.415	.085	.325	.284	.603

^aTransport speed

^bAverage mixed lay depth during transport with the definition in Section 2.4.

^cResidence time of the air mass passing over parts of highly polluted regions according to the trajectories of samples.

^dAverage air temperature.

*Correlation is significant at the 0.05 level (2-tailed).

Table S3. Dry deposition flux and increase in percentage (%)^a of aerosol particles (mg/m²/month), N_{NH4++NO3-} (mg N/m²/month) and some trace metals (mg/m²/month) for each sample on dust and comparison days

Case	Sample number	Particles (TSP)		NO ₃ ⁻ -N		NH ₄ ⁺ -N		N _{NH4++NO3-}		Fe		Cu		Pb		Zn	
		Flux	Increase	Flux	Increase	Flux	Increase	Flux	Increase	Flux	Increase	Flux	Increase	Flux	Increase	Flux	Increase
		Ratio		Ratio		Ratio		Ratio		Ratio		Ratio		Ratio		Ratio	
Case 1	20080301	7,680	174	78.42	65	23.79	59	102.21	63	508.54	169	2.06	66	0.13	44	6.83	43
	20080315	5,207	86	62.08	31	47.69	218	109.77	75	295.49	56	1.47	19	0.56	522	5.50	15
	20090316	9,254	230	54.65	15	13.27	-12	67.92	9	482.68	155	1.23	-1	0.52	478	6.52	37
	20100321	8,049	187	67.73	42	19.15	28	86.89	39	587.59	210	1.24	0	0.09	0	1.94	-59
	20110502	9,887	252	63.81	34	16.88	13	80.70	29	789.13	317	1.64	32	0.06	-33	8.34	75
	Average	8,016	186	65.3	37	24.1	61	89.5	43	532.68	181	1.53	23	0.27	202	5.83	22
Case 2	20080425	11,356	305	5.16	-89	5.14	-66	10.30	-84	424.19	124	1.72	39	0.11	22	5.34	12
	20080528	31,391	1019	1.82	-96	4.12	-73	5.94	-91	2,631.33	1290	6.02	385	0.05	-44	5.30	11
	20080529	28,053	900	0.18	-100	7.26	-52	7.44	-88	2,020.40	968	2.02	63	0.13	44	5.61	18
	20110319	12,682	352	42.47	-11	14.80	-1	57.27	-8	846.58	347	1.08	-13	0.08	-11	2.65	-44
	20110501	6,340	126	14.28	-70	8.48	-43	22.76	-64	453.61	140	2.00	61	0.02	-78	3.34	-30
	Average	17,964	540	12.8	-73	8.0	-47	20.7	-67	1,275.22	574	2.57	107	0.08	-13	4.45	-7
Case 3	20100315	12,174	334	32.24	-32	23.56	57	55.80	-11	665.76	252	6.75	444	0.20	122	3.33	-30
	20100320	65,267	2226	24.52	-48	7.91	-47	32.43	-48	4,674.81	2370	4.99	302	0.16	78	9.46	98
	20110418	10,695	281	19.92	-58	17.94	20	37.86	-40	950.51	402	6.10	392	0.17	89	3.50	-27
	Average	29,379	947	25.6	-46	16.5	10	42.0	-33	2,097.03	1008	5.9	380	0.18	96	5.4	14
ND	HDT080316	2,840		39.66		13.27		52.93		192.88		0.57		0.03		3.59	
	HDT080424	2,851		102.57		17.91		120.48		199.16		0.76		0.36		12.99	
	HDT080522	2,705		91.71		27.58		119.29		73.50		0.95		0.05		5.56	
	HDT090306	1,596		13.15		6.85		20.00		110.06		0.93		0.04		3.70	

HDT100324	3,992	27.27	4.60	31.87	449.28	4.24	0.04	3.45
HDT110308	2,573	43.59	22.76	66.35	135.26	0.99	0.04	4.63
HDT110416	3,236	18.09	8.64	26.73	197.93	0.31	0.02	0.08
HDT110523	2,658	44.40	18.69	63.08	155.97	1.19	0.11	4.15
Average	2,806	47.56	15.00	62.59	189.25	1.24	0.09	4.77

^aIncrease in percentage is calculated using the formula $(F_{Si} - F_{ANi}) / F_{ANi} \times 100\%$, where F_{Si} and F_{ANi} is the dry deposition flux of substance i in each dust day sample and average flux of comparison day samples, respectively, for those samples where the increase in percentage is positive.

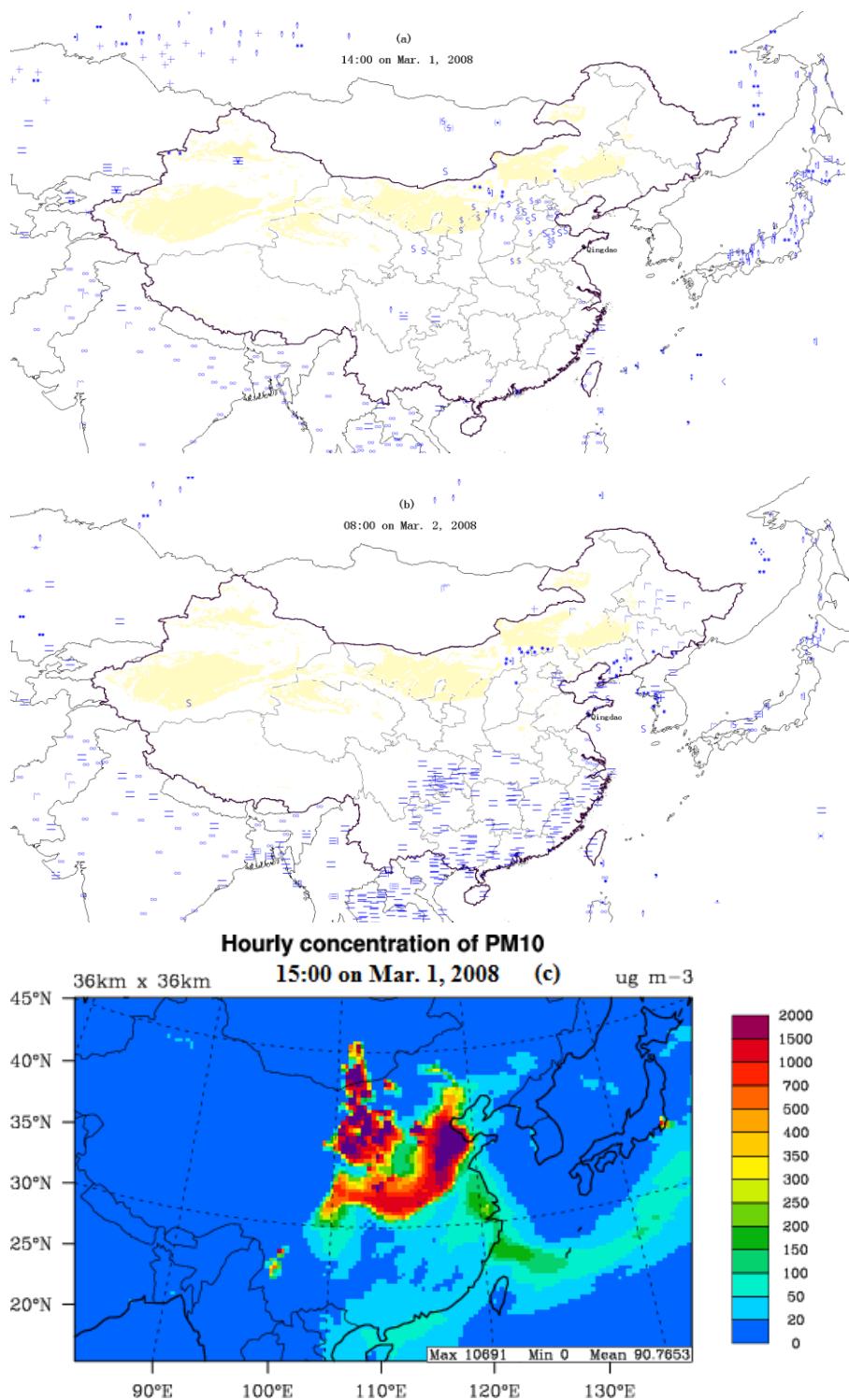


Figure S1. Dust distribution information for Sample 20080301. (a) The weather information from the MICAPS at 14:00 on Mar.1, 2008 (The symbol *S* in figure representing the dust weather). (b) The weather information from the MICAPS at 8:00 on Mar.2, 2008 (The symbol *S* in figure representing the dust weather). (c) Hourly PM10 concentration modeled by the WRF-CMAQ model at 15:00 on Mar.1, 2008.

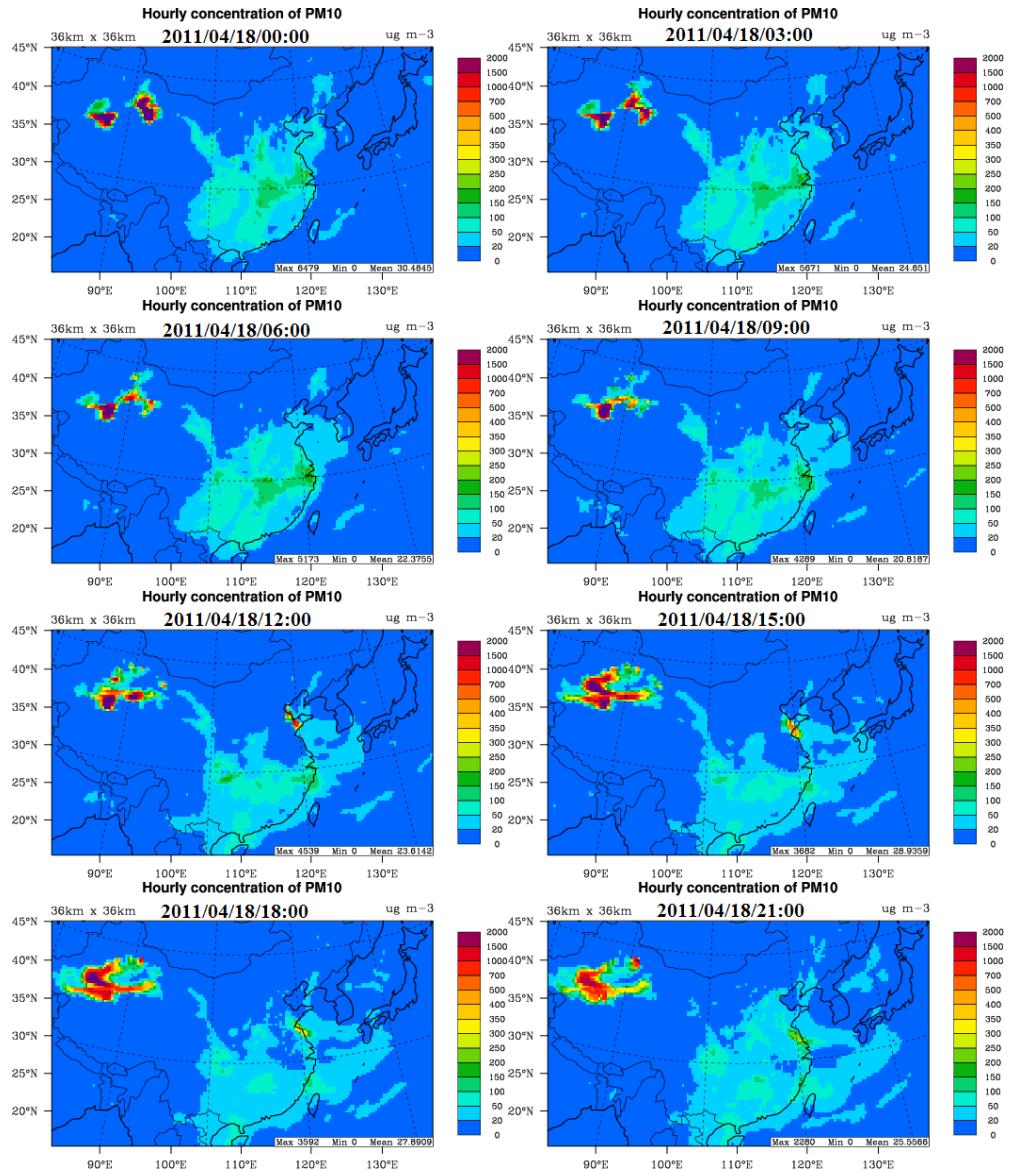


Figure S2. The modeled hourly PM₁₀ concentration over East Asia by the WRF-CMAQ model from 0:00 to 21:00 on Apr. 18, 2011.

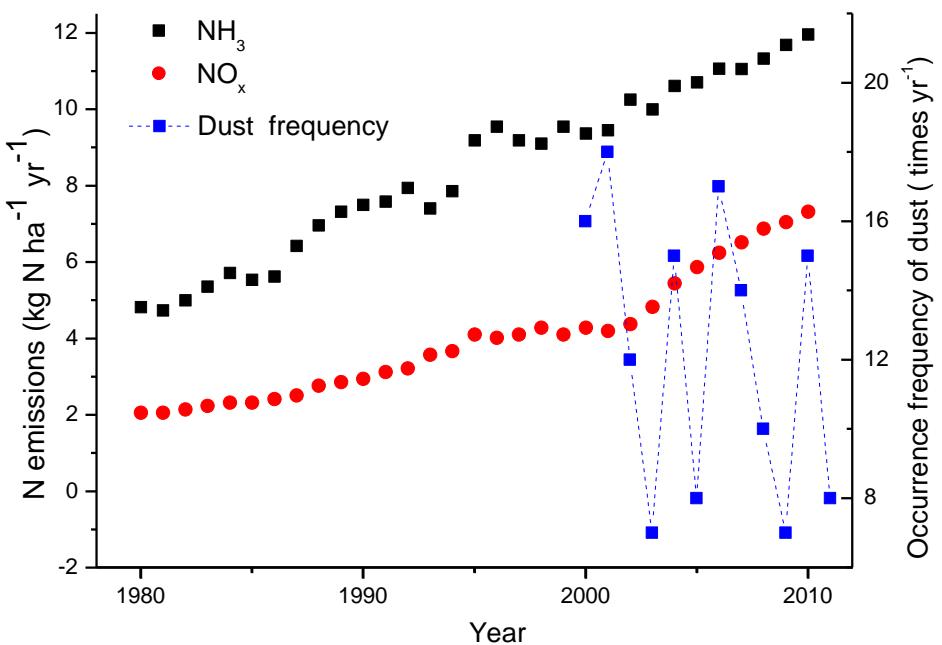


Figure S3. The NO_x and NH₃ emission and dust frequency in China in 1980-2011. (N emission during 1980-2010 from Liu et al., 2017. Dust frequency only available for 2000-2011 from CMA, 2011).

CMA: Sand-dust weather almanac 2011, China Meteorological Press, Beijing, 36-53, 2013.

Liu, L., Zhang, X., Xu, W., Liu, X., Li, Y., Lu, X., Zhang, Y., and Zhang, W.: Temporal characteristics of atmospheric ammonia and nitrogen dioxide over China based on emission data, satellite observations and atmospheric transport modeling since 1980, *Atmos. Chem. Phys.*, 17, 9365-9378, 2017.

(a)

		VAR00001	VAR00002	VAR00003	VAR00004	VAR00005	VAR00006	VAR00007	VAR00008	VAR00009	VAR00010	VAR00011	VAR00012	VAR00013	VAR00014
Spearman's rho		1.000	.582*	.308	.077	.225	.225	.341	.044	.654*	.401	.495	.182	.193	.000
Sig. (2-tailed)		.037	.306	.803	.459	.459	.255	.887	.015	.174	.085	.563	.528	1.000	
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00002		.582*	1.000	.154	.209	-.368	-.401	-.242	.286	.495	.027	-.569*	-.311	-.421	-.487
Sig. (2-tailed)		.037	.	.616	.494	.216	.174	.426	.344	.086	.929	.042	.301	.152	.091
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00003		.308	.154	1.000	.698**	.533	.588*	.555*	.731**	.527	.209	-.358	.512	.083	.072
Sig. (2-tailed)		.306	.616	.	.008	.061	.035	.049	.005	.064	.494	.230	.074	.789	.816
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00004		.077	.209	.698**	1.000	.148	.319	.297	.852**	.264	-.038	-.432	.366	-.311	-.206
Sig. (2-tailed)		.803	.494	.008	.	.629	.289	.325	.000	.384	.901	.141	.219	.301	.499
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00005		.225	-.368	.533	.148	1.000	.775**	.599*	.220	.286	.275	.069	.514	.492	.525
Sig. (2-tailed)		.459	.216	.061	.629	.	.002	.031	.471	.344	.364	.823	.072	.087	.065
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00006		.225	-.401	.588*	.319	.775**	1.000	.879*	.159	.440	.319	.047	.751**	.107	.388
Sig. (2-tailed)		.459	.174	.035	.289	.002	.	.000	.603	.133	.289	.879	.003	.727	.190
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00007		.341	-.242	.555*	.297	.599*	.879**	1.000	.082	.637*	.577*	-.050	.798**	.143	.424
Sig. (2-tailed)		.255	.426	.049	.325	.031	.000	.	.789	.019	.039	.872	.001	.641	.149
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00008		.044	.288	.731**	.852**	.220	.159	.082	1.000	.220	-.126	-.314	.204	-.044	-.182
Sig. (2-tailed)		.887	.344	.005	.000	.471	.603	.789	.	.471	.681	.297	.505	.886	.553
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00009		.654*	.495	.527	.264	.286	.440	.637*	.220	1.000	.599*	-.223	.437	-.008	.118
Sig. (2-tailed)		.015	.086	.064	.384	.344	.133	.019	.471	.	.031	.464	.135	.979	.700
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00010		.401	.027	.209	-.038	.275	.319	.577*	-.126	.599*	1.000	.	.228	.558*	.465
Sig. (2-tailed)		.174	.929	.494	.901	.364	.289	.039	.681	.031	.	.453	.047	.109	.027
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00011		-.495	-.569*	-.358	-.432	.069	.047	-.050	-.314	-.223	.228	1.000	-.025	.438	.267
Sig. (2-tailed)		.085	.042	.230	.141	.823	.879	.872	.297	.464	.453	.	.936	.134	.377
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00012		.182	-.311	.512	.366	.514	.751**	.798**	.204	.437	.558*	-.025	1.000	.322	.675*
Sig. (2-tailed)		.553	.301	.074	.219	.072	.003	.001	.505	.135	.047	.936	.	.283	.011
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00013		-.193	-.421	.083	-.311	.492	.107	.143	-.044	-.008	.465	.438	.322	1.000	.802**
Sig. (2-tailed)		.528	.152	.789	.301	.067	.727	.641	.886	.979	.109	.134	.283	.	.001
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13
VAR00014		.000	-.487	.072	-.206	.525	.388	.424	-.182	.118	.608*	.267	.675*	.802**	1.000
Sig. (2-tailed)		1.000	.091	.816	.499	.065	.190	.149	.553	.700	.027	.377	.011	.001	.
N		13	13	13	13	13	13	13	13	13	13	13	13	13	13

(b)

		VAR00001	VAR00002	VAR00003	VAR00004	VAR00005	VAR00006	VAR00007	VAR00008	VAR00009	VAR00010	VAR00011	VAR00012	VAR00013	VAR00014
Spearman's rho		1.000	-.400	-.900*	-.600	.308	.000	-.300	-.700	-.400	.300	.900*	-.300	.100	.400
Sig. (2-tailed)		.505	.037	.285	.104	.624	1.000	.624	.188	.505	.624	.037	.624	.873	.505
N		5	5	5	5	5	5	5	5	5	5	5	5	5	5
VAR00002		-.400	1.000	.300	.800	-.900*	.100	.900*	.500	1.000**	.400	.000	.600	-.500	-.300
Sig. (2-tailed)		.505	.	.624	.104	.037	.873	.037	.391	.	.505	1.000	.285	.391	.624
N		5	5	5	5	5	5	5	5	5	5	5	5	5	5
VAR00003		-.900*	.300	1.000	.700	-.100	.100	.400	.900*	.300	-.400	-.800	-.100	-.200	-.700
Sig. (2-tailed)		.037	.624	.	.188	.873	.873	.505	.037	.624	.505	.104	.873	.747	.188
N		5	5	5	5	5	5	5	5	5	5	5	5	5	5
VAR00004		-.600	.800	.700	1.000	-.500	.400	.900*	.900*	.800	-.100	-.300	.100	-.700	-.800
Sig. (2-tailed)		.285	.104	.188	.	.391	.505	.037	.037	.104	.873	.624	.873	.188	.104
N		5	5	5	5	5	5	5	5	5	5	5	5	5	5
VAR00005		.300	-.900*	-.100	-.500	1.000	.300	-.700	-.200	-.900*	-.700	1.000	-.100	-.800	-.100
Sig. (2-tailed)		.624	.037	.873	.873	.505	.624	.	.747	.624	.873	.037	.873	.873	.873
N		5	5	5	5	5	5	5	5	5	5	5	5	5	5
VAR00006		.000	.100	.100	.400	.300	1.000	.200	.300	.100	.700	-.700	-.100	-.300	-.900*
Sig. (2-tailed)		.1000	.873	.873	.505	.624	.	.747	.624	.873	.037	.873	.873	.873	.873
N		5	5	5	5	5	5	5	5	5	5	5	5	5	5
VAR00007		-.300	-.900*	.400	-.900*	-.700	.200	1.000	.700	.900*	.300	.100	.200	-.600	-.600
Sig. (2-tailed)		.624	.037	.505	.037	.188	.747	.	.188	.037	.624	.873	.747	.285	.285
N		5	5	5	5	5	5	5	5	5	5	5	5	5	5
VAR00008		-.700	.500	.900*	-.900*	-.200	.300	.700	1.000	.500	-.300	-.500	-.200	-.500	-.900*
Sig. (2-tailed)		.188	.391	.037	.037	.747	.624	.188	.	.391	.624	.391	.747	.391	.037
N		5	5	5	5	5	5	5	5	5	5	5	5	5	5
VAR00009		-.400	1.000**	.300	.800	-.900*	.100	.900*	.500	1.000	.400	.000	.600	-.500	-.500
Sig. (2-tailed)		.505	.624	.104	.037	.873	.	.391	.505	1.000	.505	1.000	.285	.391	.624
N		5	5	5	5	5	5	5	5	5	5	5	5	5	5
VAR00010		.300	.400	-.400	-.100	-.700	-.700	1.000	.300	-.300	.400	1.000	.600	.500	.500
Sig. (2-tailed)		.624	.505	.505	.873	.188	.624	.	.624	.505	.505	.285	.391	.505	.391
N		5	5	5	5</td										

		VAR00001	VAR00002	VAR00003	VAR00004	VAR00005	VAR00006	VAR00007	VAR00008	VAR00009	VAR00010	VAR00011	VAR00012	VAR00013	VAR00014		
Spearman's rho	VAR00001	Correlation Coefficient	1.000	.200	1.000*	.300	.700	.500	.700	.900*	.100	.-300	.200	.600	.700		
		Sig. (2-tailed)		.747		.624	.188	.391	.391	.188	.037	.873	.624	.747	.285	.188	
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00002	Correlation Coefficient	.200	1.000	.200	.000	-.200	-.600	.300	-.100	-.400	-.300	-.800	.100	.100	-.200	
		Sig. (2-tailed)		.747		.747	1.000	.747	.285	.873	.055	.873	.624	.104	.873	.747	
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00003	Correlation Coefficient	1.000*	.200	1.000	.300	.700	.500	.700	.900*	.100	.-300	.200	.600	.700		
		Sig. (2-tailed)		.747		.624	.188	.391	.391	.188	.037	.873	.624	.747	.285	.188	
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00004	Correlation Coefficient	.300	.000	.300	1.000	.200	.100	.100	.700	.100	-.600	-.700	.300	-.100	.200	
		Sig. (2-tailed)		.624		.624	.	.747	.873	.188	.873	.285	.188	.624	.873	.747	
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00005	Correlation Coefficient	.700	-.200	.700	.200	1.000	.900*	.900*	.700	.900*	.600	.300	.700	.900*	1.000**	
		Sig. (2-tailed)		.188		.747	.188	.747	.	.037	.188	.037	.285	.624	.188	.037	.
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00006	Correlation Coefficient	.500	-.600	.500	.100	.900*	1.000	1.000*	.400	.800	.700	.400	.900*	.700	.900*	
		Sig. (2-tailed)		.391		.285	.391	.873	.037	.	.505	.104	.188	.505	.037	.188	.037
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00007	Correlation Coefficient	.500	-.600	.500	.100	.900*	1.000*	1.000*	.400	.800	.700	.400	.900*	.700	.900*	
		Sig. (2-tailed)		.391		.285	.391	.873	.037	.	.505	.104	.188	.505	.037	.188	.037
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00008	Correlation Coefficient	.700	.300	.700	.700	.700	.400	.400	1.000	.600	-.100	-.300	.300	.600	.700	
		Sig. (2-tailed)		.188		.624	.188	.188	.188	.505	.	.285	.873	.624	.285	.188	
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00009	Correlation Coefficient	.900*	-.100	.900*	.100	.900*	.800	.800	.600	1.000	.500	.100	.500	.800	.900*	
		Sig. (2-tailed)		.037		.873	.037	.873	.037	.104	.104	.285	.	.873	.391	.104	.037
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00010	Correlation Coefficient	.100	-.400	.100	-.600	.600	.700	.700	-.100	.500	1.000	.900*	.500	.700	.600	
		Sig. (2-tailed)		.873		.505	.873	.285	.285	.188	.873	.391	.	.037	.391	.188	.285
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00011	Correlation Coefficient	-.300	-.300	-.300	-.700	.300	.400	.400	-.300	.100	.900*	1.000	.100	.300	.500	.300
		Sig. (2-tailed)		.624		.624	.188	.824	.505	.505	.624	.873	.037	.624	.391	.624	
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00012	Correlation Coefficient	.200	-.800	.200	.300	.700	.900*	.900*	.300	.500	.500	.300	1.000	.400	.700	
		Sig. (2-tailed)		.747		.104	.747	.624	.188	.037	.037	.624	.391	.391	.624	.505	.188
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00013	Correlation Coefficient	.600	.100	.600	-.100	.900*	.700	.700	.600	.800	.700	.500	.400	1.000	.900*	
		Sig. (2-tailed)		.285		.873	.285	.873	.037	.188	.285	.104	.188	.391	.505	.037	.
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	
	VAR00014	Correlation Coefficient	.700	-.200	.700	.200	1.000*	.900*	.900*	.700	.900*	.600	.300	.700	.900*	1.000	
		Sig. (2-tailed)		.188		.747	.188	.747	.	.037	.037	.188	.037	.285	.624	.188	.
		N	5	5	5	5	5	5	5	5	5	5	5	5	5	5	

Figure S4. The Spearman correlation coefficient of nitrate and ammonium with other ions in aerosols samples for all samples (a), Category 1 (b) and Category 2 (c) . (Var00001, VAR00002, VAR00003, VAR00004,..., AR00014 Successively representing NO_3^- -N, NH_4^+ -N, SO_4^{2-} -S, Cl^- , Ca^{2+} , Mg^{2+} , K^+ , Na^+ , Pb , Zn , Cu , Ca , Fe , Al , respectively.)

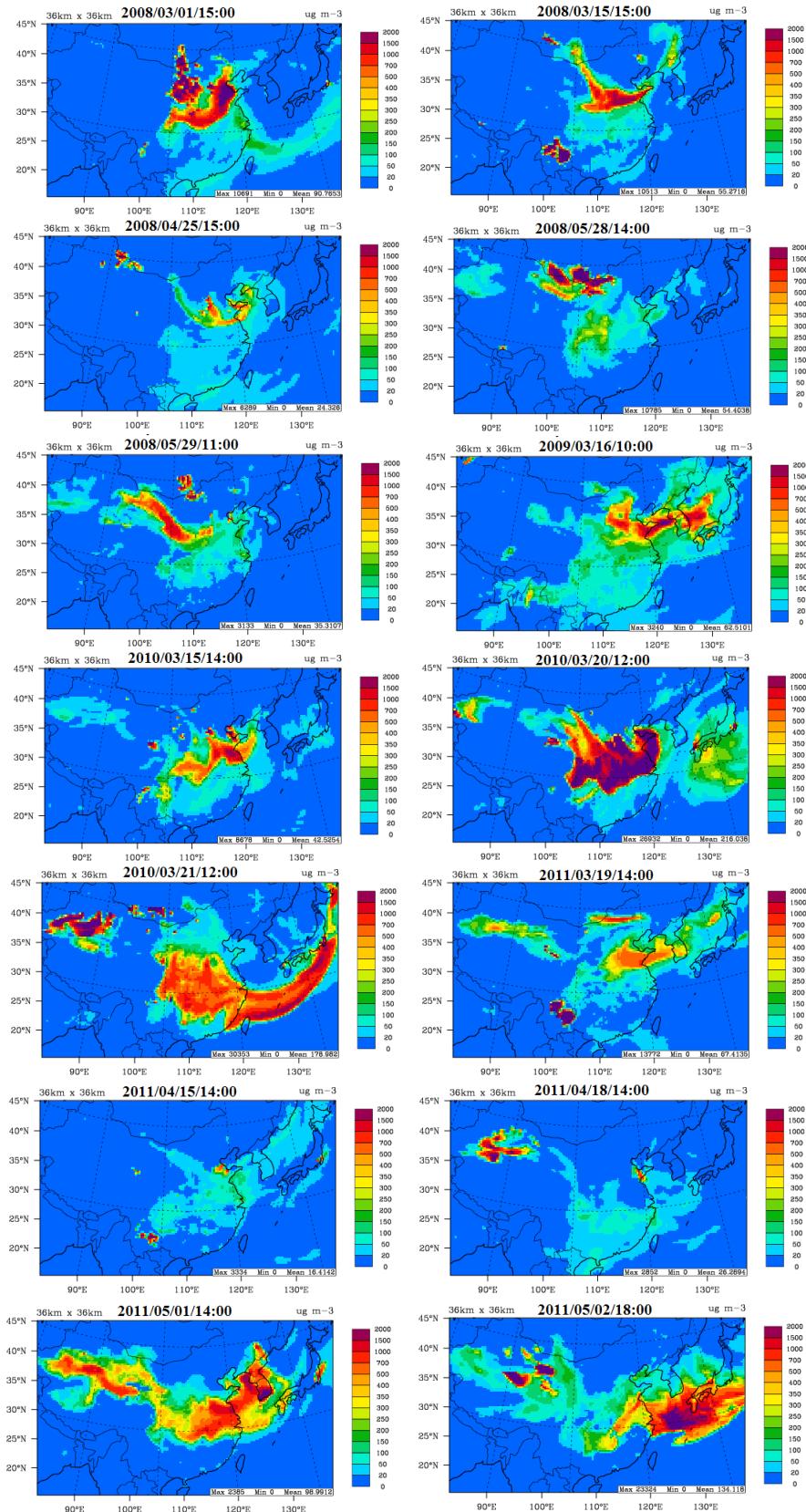


Figure S5. Hourly PM₁₀ concentration over East Asia modeled by the WRF-CMAQ model for each dust sampling day from 2008 to 2011 (the PM₁₀ concentration in the middle of each sampling duration only showed).

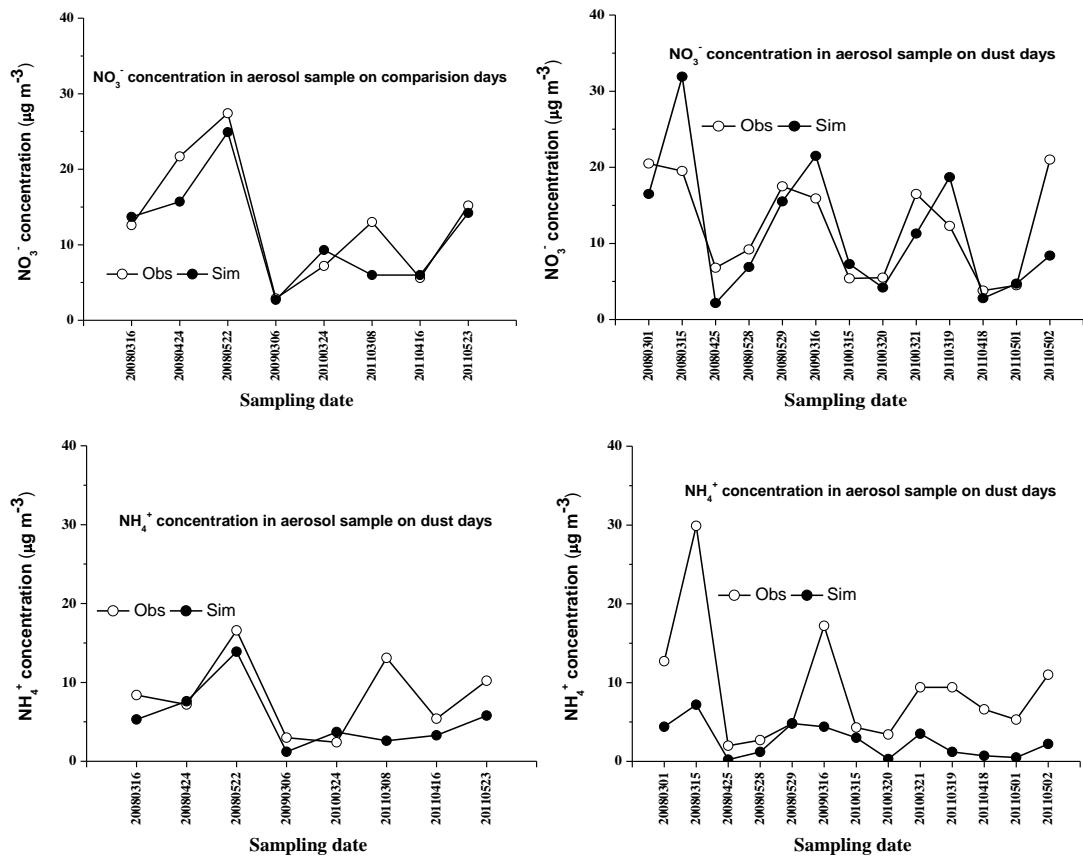


Figure S6. The comparison between simulated and observed NO_3^- and NH_4^+ in aerosol samples on dust and comparison days in Qingdao in 2008 to 2011.