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Size-resolved source apportionment of particulate matter in urban Beijing during haze and non-haze episodes

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Table S1 Calculation methods of main components used in mass closure study

	Calculation Methods
CM	$CM=SiO_2+Al_2O_3+CaO+Fe_2O_3+K_2O+Na_2O+MgO=1.89[Al]+1.66[Mg]_n+1.21[K]+1.40[Ca]_n+1.43[Fe]_n+1.35[Na - ss\text{-}Na^+]+2.14[Si]$
OM	$OM=1.4\times OC$
HM	HM=Cu+Pb+Zn+Cd+As+Cr+V+Mn
SS	$SS=[Na^+]+[SS\text{-}Cl^-]+[SS\text{-}Mg^{2+}]+[SS\text{-}Ca^{2+}]+[SS\text{-}K^+]+[SS\text{-}SO_4^{2-}]=3.246[Na^+]$
SNA	$SNA=[NSS\text{-}SO_4^{2-}]+[NO_3^-]+[NH_4^+]$
UM	UM=PM-CM-OM-HM-SS-SIA-EC-LW

The calculation method for secondary inorganic aerosol and sea salt can be found in (Terzi et al., 2010). Moreover, $[NSS\text{-}SO_4^{2-}]$ refers to water-soluble SO_4^{2-} apart from sea salt; $[SS\text{-}Na^+]$ refers to water-soluble Na^+ in sea salt. The calculation method for crustal materials can be found in (Hueglin et al., 2005); $[Si_n]$, $[Fe_n]$, $[Ca_n]$ and $[Mg_n]$ were calculated based on the ratio to Al in the crustal materials. Liquid water (LW) was calculated using an aerosol thermodynamic model of E-AIM II(Extended Aerosol Inorganic Model, http://www.aim.env.uea.ac.uk/aim/aim.php)(Clegg et al., 1998)

Table S2 Concentrations of different chemical compositions in size-resolved particles during entire sampling period (annual) and four seasons ($\mu g\ m^{-3}$).

	Annual				Spring				Summer				Autumn				Winter			
	Clear		Haze		Clear		Haze		Clear		Haze		Clear		Haze		Clear		Haze	
Size	$PM_{2.1}$	$PM_{2.1\text{-}9}$	$PM_{2.1}$	$PM_{2.1\text{-}9}$	$PM_{2.1}$	$PM_{2.1\text{-}9}$	$PM_{2.1}$	$PM_{2.1\text{-}9}$	$PM_{2.1}$	$PM_{2.1\text{-}9}$	$PM_{2.1}$	$PM_{2.1\text{-}9}$	$PM_{2.1}$	$PM_{2.1\text{-}9}$	$PM_{2.1}$	$PM_{2.1\text{-}9}$	$PM_{2.1}$	$PM_{2.1\text{-}9}$	$PM_{2.1}$	$PM_{2.1\text{-}9}$
Mass	32.6	50.5	86.1	72.6	41.0	70.5	74.7	80.2	21.8	34.2	48.8	65.0	37.1	46.4	83.1	76.2	20.4	42.6	114.2	67.7
OC	8.8	5.9	19.2	9.7	14.9	12.6	17.4	10.9	16.6	13.8	19.2	15.5	5.9	2.9	18.3	9.9	5.4	1.5	21.3	5.8
EC	1.2	0.4	2.6	1.0	1.1	0.3	1.5	0.7	1.2	0.5	1.3	0.7	1.5	0.5	1.9	1.2	0.8	0.1	4.5	1.0
Na^+	0.4	0.4	0.6	0.8	0.4	0.4	0.7	0.8	0.2	0.2	0.3	0.5	0.4	0.4	0.6	0.6	0.5	0.5	0.7	1.1
NH_4^+	2.0	0.2	8.0	1.1	3.5	0.4	12.0	1.0	1.2	0.1	3.7	0.3	1.6	0.2	6.3	0.7	1.5	0.2	9.2	1.9
K^+	0.4	0.2	0.9	0.3	0.6	0.3	1.2	0.6	0.1	0.0	0.4	0.1	0.3	0.1	0.7	0.1	0.6	0.3	1.3	0.5
Mg^{2+}	0.2	0.3	0.2	0.5	0.2	0.4	0.2	0.5	0.2	0.3	0.1	0.4	0.2	0.4	0.2	0.5	0.2	0.3	0.2	0.4
Ca^{2+}	1.0	3.1	1.0	3.9	1.0	3.8	1.0	5.4	0.6	2.5	0.7	3.1	1.0	3.3	1.1	4.2	1.1	2.2	1.1	2.9

Cl^-	0.9	0.4	2.2	1.2	0.8	0.8	2.5	1.7	0.1	0.1	0.1	0.4	1.1	0.3	1.4	0.5	0.9	0.4	3.8	1.9
NO_3^-	2.1	1.3	11.4	4.1	3.4	2.9	11.8	5.3	0.3	1.2	2.0	3.2	2.0	0.6	8.4	3.0	1.5	0.7	18.7	4.9
SO_4^{2-}	2.9	0.9	13.1	3.0	7.3	1.5	14.9	3.8	2.4	0.3	8.3	1.1	1.1	0.7	11.6	2.5	2.0	0.5	15.7	3.9
Na	1.7	1.4	1.9	1.5	1.9	1.7	1.9	1.5	1.7	0.8	1.7	1.2	1.6	1.3	1.9	1.3	1.6	1.5	1.9	1.7
Mg	0.5	1.1	0.4	1.4	0.7	2.2	0.5	2.0	0.5	0.8	0.5	1.4	0.5	0.8	0.4	1.3	0.3	0.7	0.3	1.0
Al	0.7	1.5	0.6	1.5	0.9	3.3	0.7	2.4	0.7	0.8	0.5	1.0	0.7	1.2	0.7	1.4	0.5	1.0	0.6	1.1
K	0.4	0.5	0.9	0.8	0.7	1.4	1.0	1.2	0.3	0.3	0.5	0.6	0.3	0.3	0.9	0.8	0.2	0.4	1.0	0.5
Ca	3.5	10.7	3.4	10.6	5.6	29.5	3.5	17.7	4.9	7.4	5.1	13.0	2.8	6.8	3.6	9.3	2.3	5.5	2.3	5.9
Mn	0.03	0.04	0.04	0.05	0.04	0.10	0.05	0.07	0.03	0.02	0.03	0.03	0.03	0.03	0.05	0.04	0.01	0.03	0.04	0.05
Fe	1.13	2.48	1.09	2.93	2.15	6.48	1.34	4.07	1.81	1.66	1.04	2.24	0.92	1.65	1.45	3.08	0.35	1.45	0.61	2.36
Co	0.001	0.001	0.001	0.001	0.001	0.002	0.001	0.002	0.000	0.001	0.000	0.001	0.000	0.001	0.001	0.001	0.000	0.001	0.001	0.001
Ni	0.013	0.016	0.013	0.013	0.011	0.014	0.011	0.013	0.018	0.022	0.014	0.008	0.011	0.017	0.014	0.013	0.015	0.013	0.013	0.015
Cu	0.019	0.020	0.034	0.022	0.029	0.030	0.034	0.022	0.012	0.013	0.016	0.014	0.018	0.021	0.037	0.024	0.015	0.015	0.038	0.024
Zn	0.150	0.076	0.253	0.111	0.291	0.126	0.320	0.161	0.203	0.092	0.186	0.093	0.118	0.071	0.279	0.098	0.089	0.047	0.220	0.101
Mo	0.001	0.001	0.002	0.002	0.002	0.001	0.001	0.001	0.001	0.002	0.002	0.001	0.001	0.001	0.002	0.002	0.002	0.001	0.003	0.002
Cd	0.001	0.000	0.002	0.000	0.002	0.000	0.003	0.001	0.001	0.000	0.001	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.002	0.000
Ba	0.016	0.039	0.017	0.050	0.022	0.080	0.016	0.066	0.012	0.026	0.013	0.035	0.015	0.032	0.019	0.052	0.014	0.029	0.018	0.045
Tl	0.001	0.000	0.001	0.000	0.001	0.000	0.002	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.001	0.000
Pb	0.047	0.012	0.134	0.029	0.114	0.032	0.209	0.056	0.029	0.006	0.083	0.014	0.033	0.009	0.105	0.017	0.025	0.006	0.136	0.030
Th	0.000	0.000	0.000	0.001	0.000	0.001	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001
U	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Table S3 The correlation coefficients between chemical species in different size fractions and visibility

Size	OC	EC	Na^+	MH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
<0.43	-0.316	-0.192	-0.158	**-0.393**	-0.321	-0.174	-0.199	-0.355	**-0.350**	**-0.303**
0.43-0.65	**-0.371**	-0.189	-0.113	**-0.506**	**-0.380**	-0.215	-0.248	**-0.414**	**-0.445**	**-0.590**
0.65-1.1	**-0.485**	-0.119	-0.200	**-0.418**	**-0.384**	-0.218	-0.282	**-0.377**	**-0.470**	**-0.582**
1.1-2.1	-0.296	-0.173	-0.180	**-0.414**	-0.356	-0.199	-0.279	-0.308	**-0.359**	**-0.388**
2.1-3.3	-0.234	-0.196	-0.196	-0.217	-0.295	-0.238	-0.358	**-0.319**	**-0.340**	**-0.302**
3.3-4.7	-0.207	-0.209	-0.139	-0.218	-0.255	-0.196	-0.421	-0.195	**-0.433**	-0.255
4.7-5.8	-0.196	-0.183	-0.206	-0.242	-0.189	-0.245	-0.481	-0.258	**-0.406**	-0.232
5.8-9	-0.354	-0.123	-0.218	-0.318	-0.193	**-0.426**	**-0.520**	**-0.311**	**-0.507**	**-0.461**
>9	-0.477	-0.145	-0.185	-0.245	-0.183	-0.319	-0.388	-0.195	**-0.425**	**-0.346**

Fig. S1 Map of the site location in Beijing (A was the sampling site, http://www.google.cn/maps).

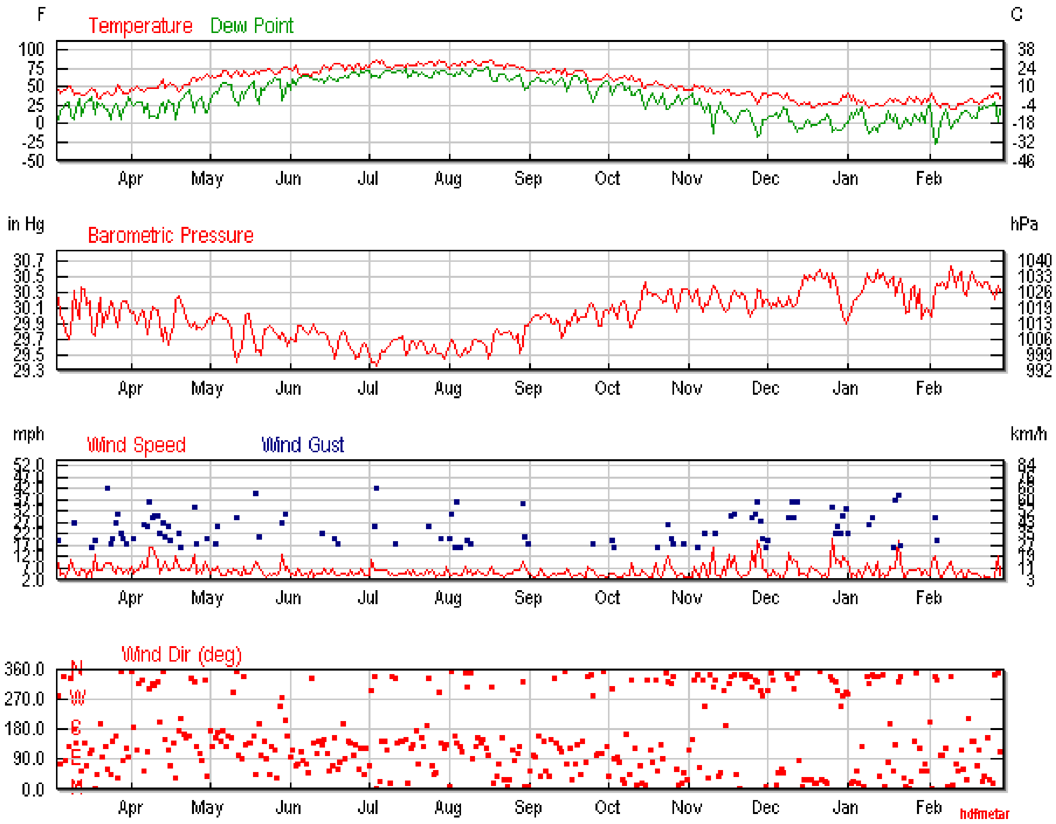
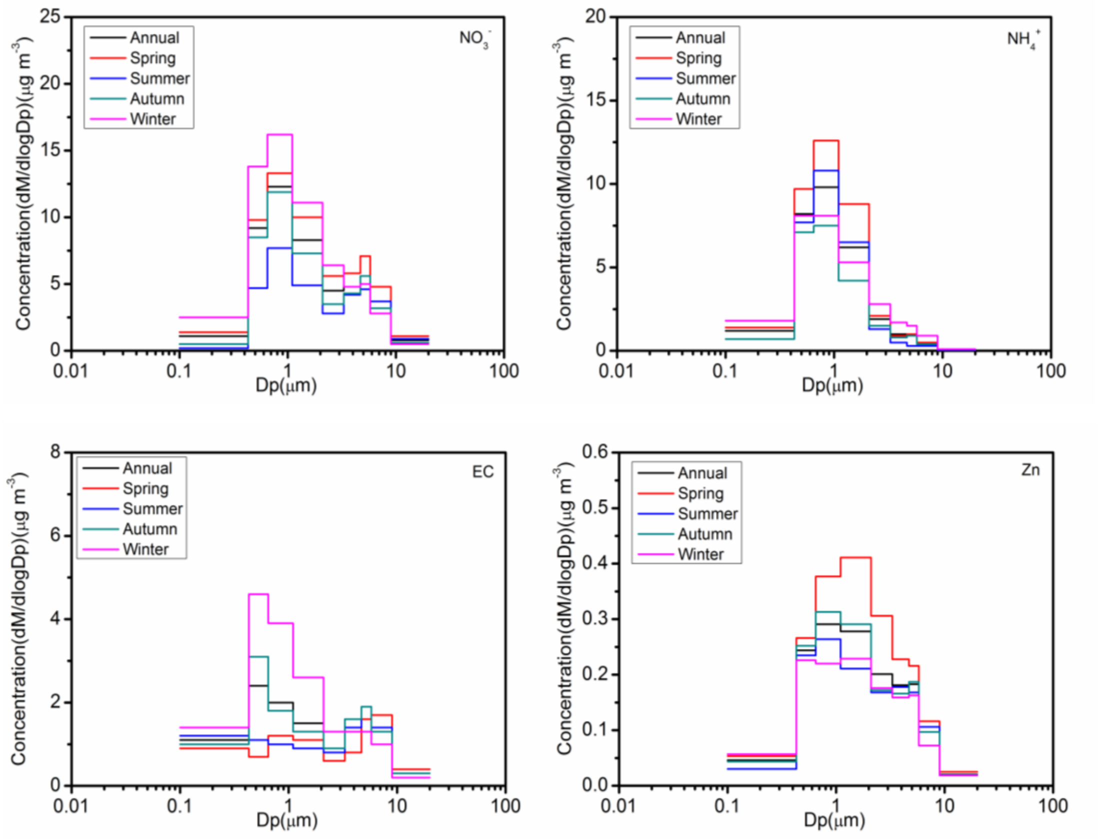
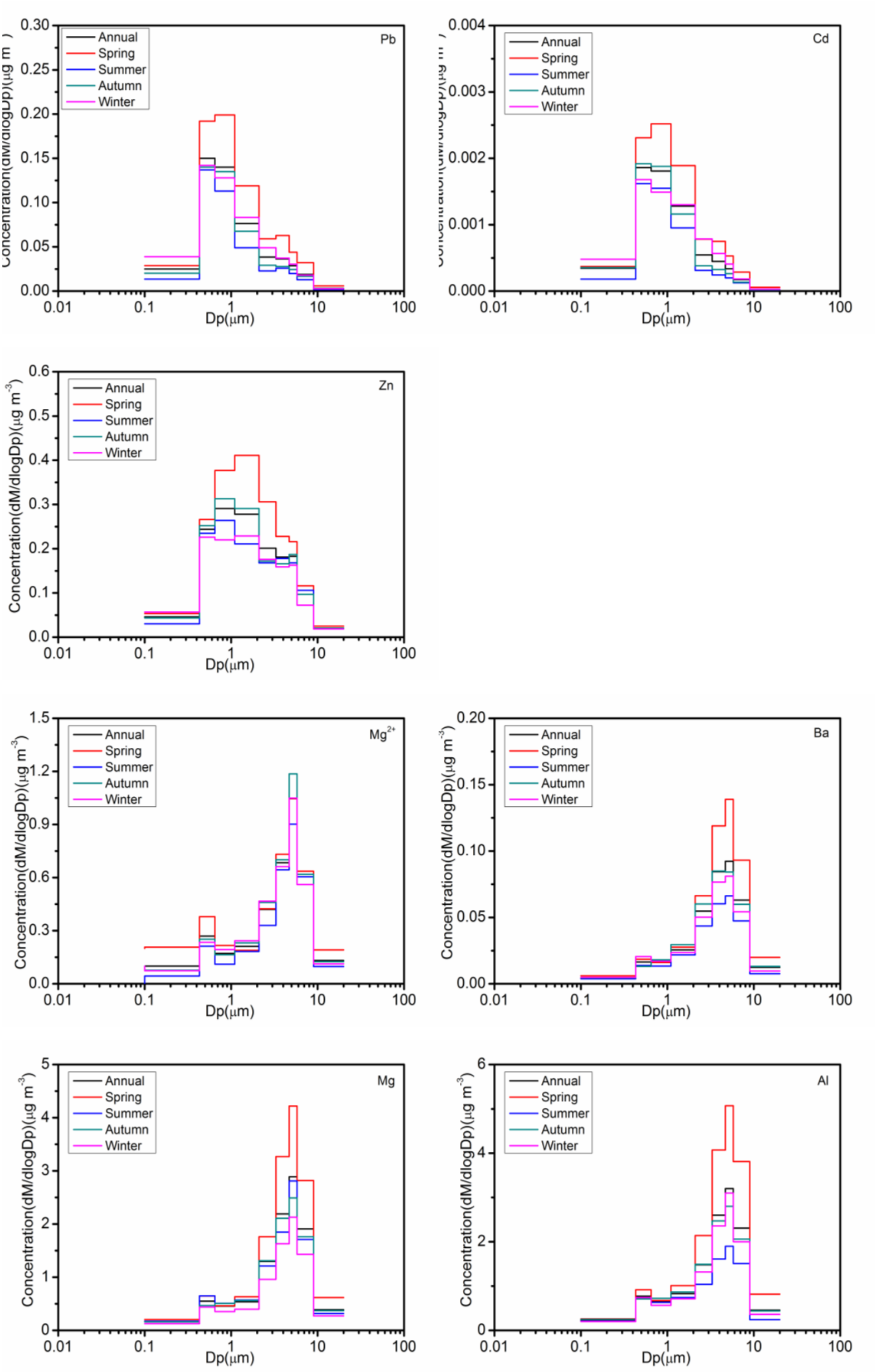
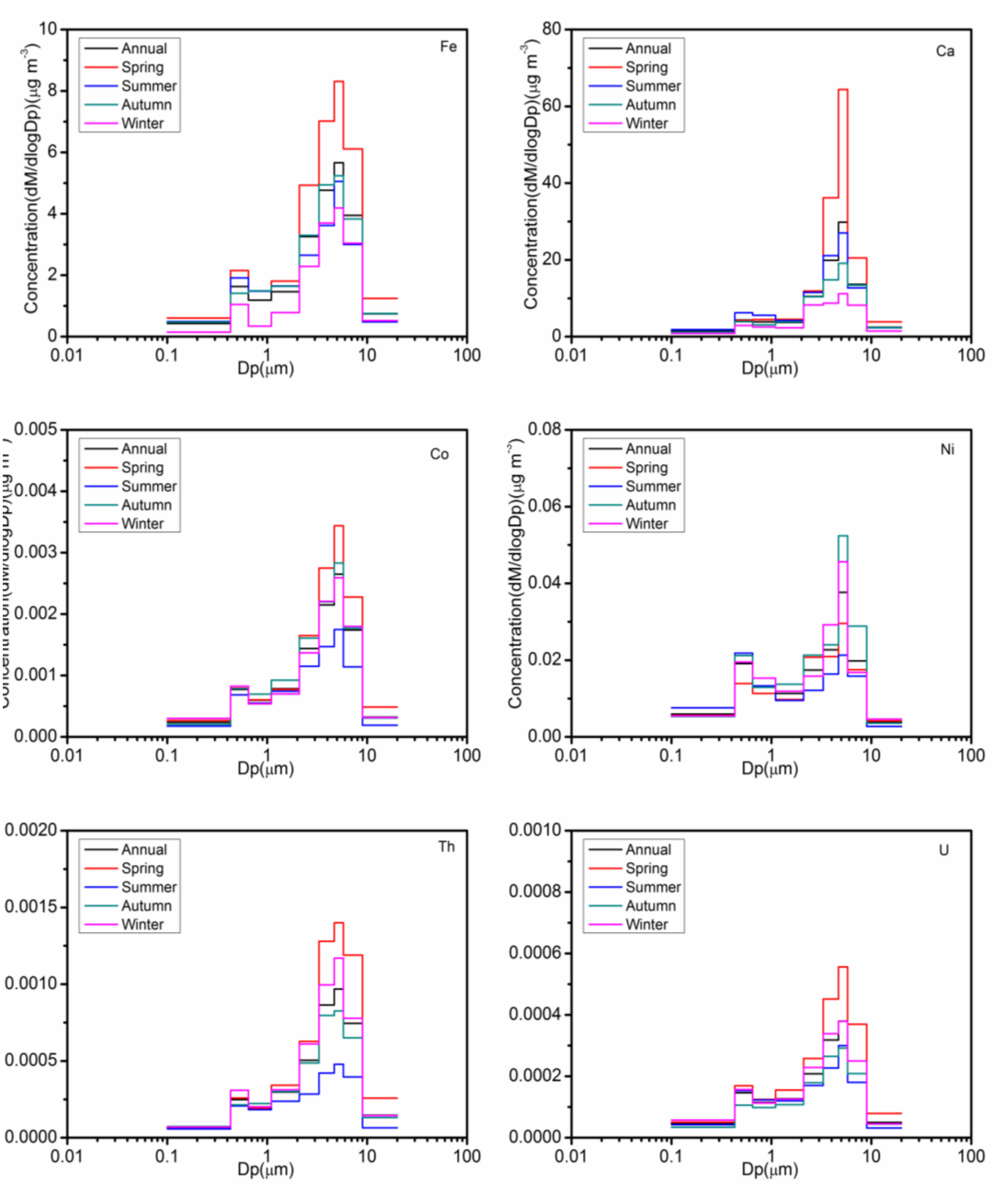


Fig. S2 Meteorological parameters in Beijing from March 1, 2013, to February 28, 2014 (http://english.wunderground.com)




Pb
Cd
Zn
Mg^{2+}
Ba
Mg
Al
Annual
Spring
Summer
Autumn
Winter
Concentration(dM/dlogDp)(μg m^{-3})
Dp(μm)



Fe
Annual
Spring
Summer
Autumn
Winter
Concentration(dM/dlogDp)(μg m-3)
Dp(μm)
Ca
Annual
Spring
Summer
Autumn
Winter
Concentration(dM/dlogDp)(μg m-3)
Dp(μm)
Co
Annual
Spring
Summer
Autumn
Winter
Dp(μm)
Ni
Annual
Spring
Summer
Autumn
Winter
Concentration(dM/dlogDp)(μg m-3)
Dp(μm)
Th
Annual
Spring
Summer
Autumn
Winter
Dp(μm)
U
Annual
Spring
Summer
Autumn
Winter
Dp(μm)

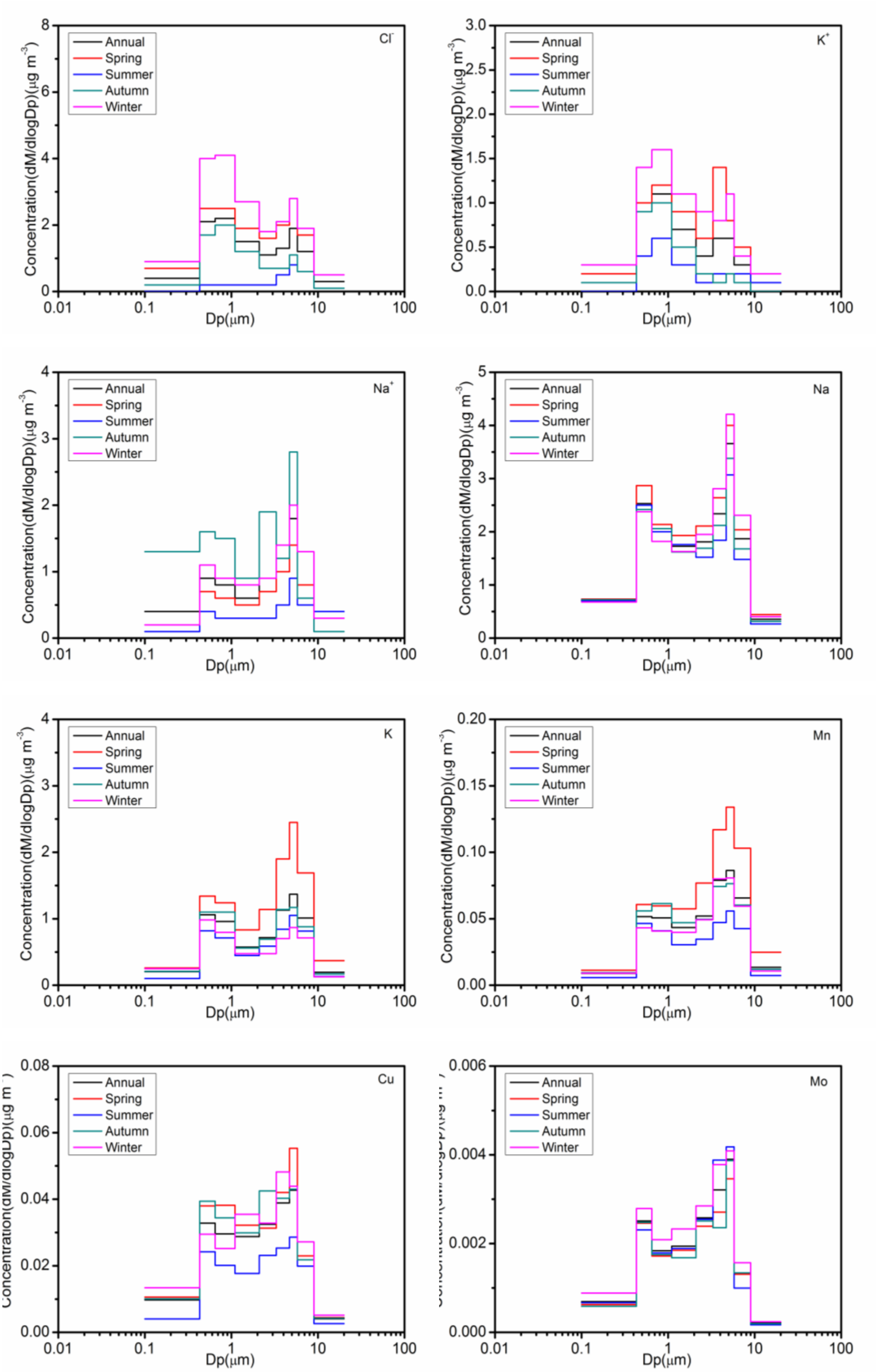


Fig. S3 Size distributions of chemical species in different seasons.

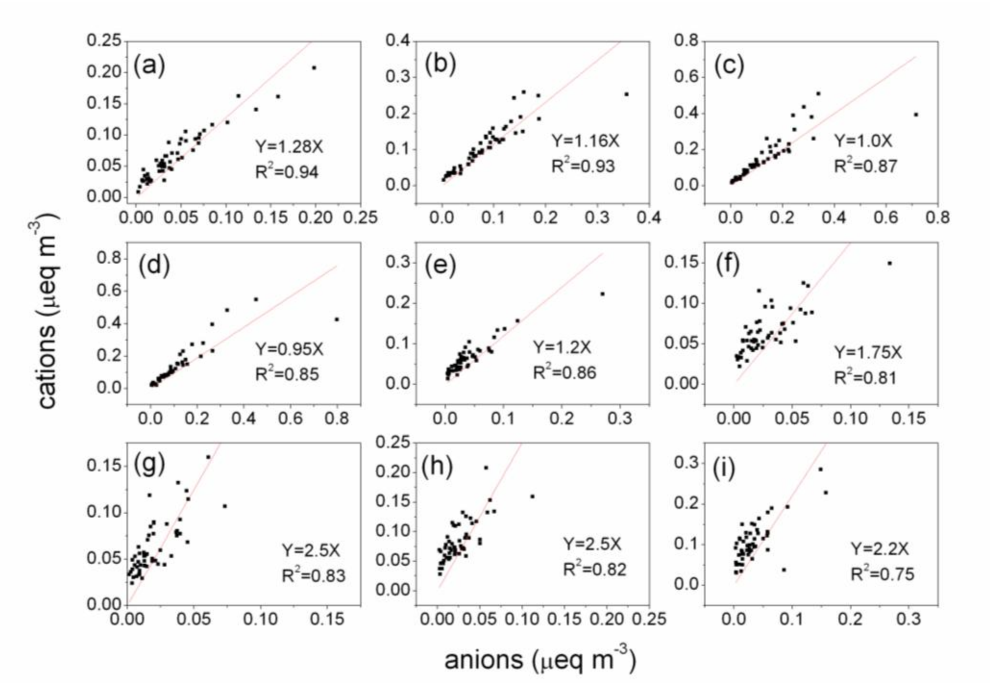


Fig. S4 Ion balance for different size fractions: (a) <0.43 μm; (b) 0.43-0.65μm; (c) 0.65-1.1μm; (d) 1.1-2.1 μm; (e) 2.1-3.3 μm; (f) 3.3-4.7 μm; (g) 4.7-5.8 μm; (h) 5.8-9 μm; (i) >9μm.

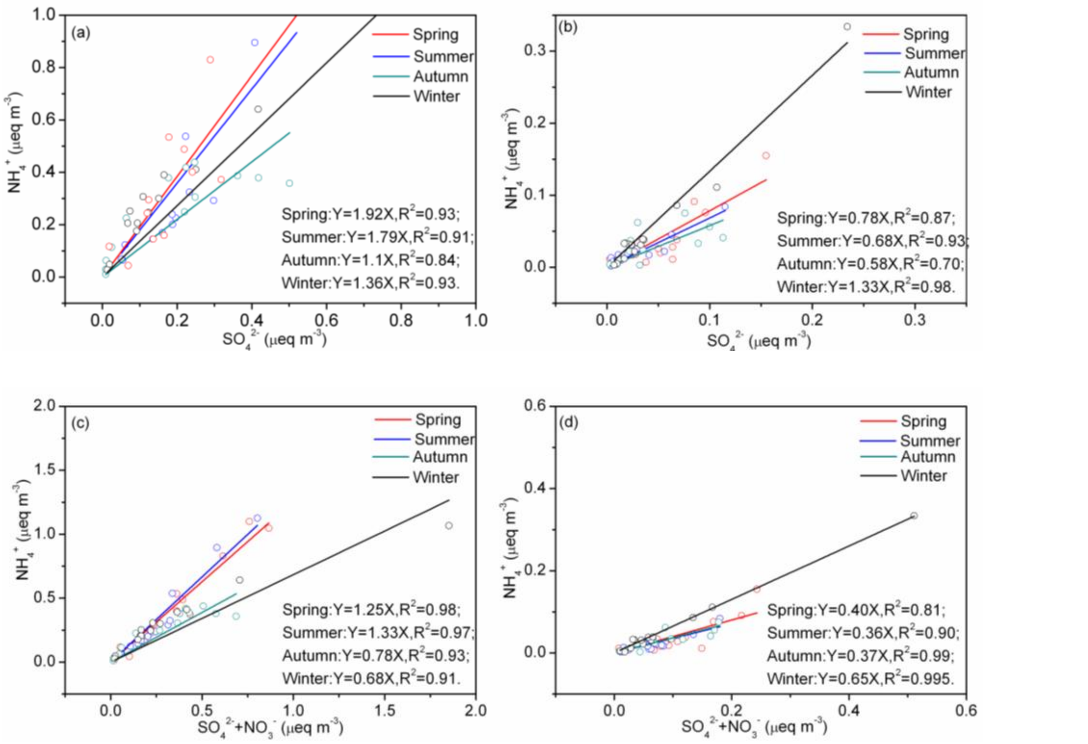


Fig. S5 Correlations between certain cations and anions in both fine and coarse particles during different seasons: (a) NH_4^+ and SO_4^{2-} in fine particles; (b) NH_4^+ and SO_4^{2-} in coarse particles; (c) $NH_4^+$to [NO_3^- + SO_4^{2-}] in fine particles;; (d) $NH_4^+$to [NO_3^- + SO_4^{2-}] in coarse particles.

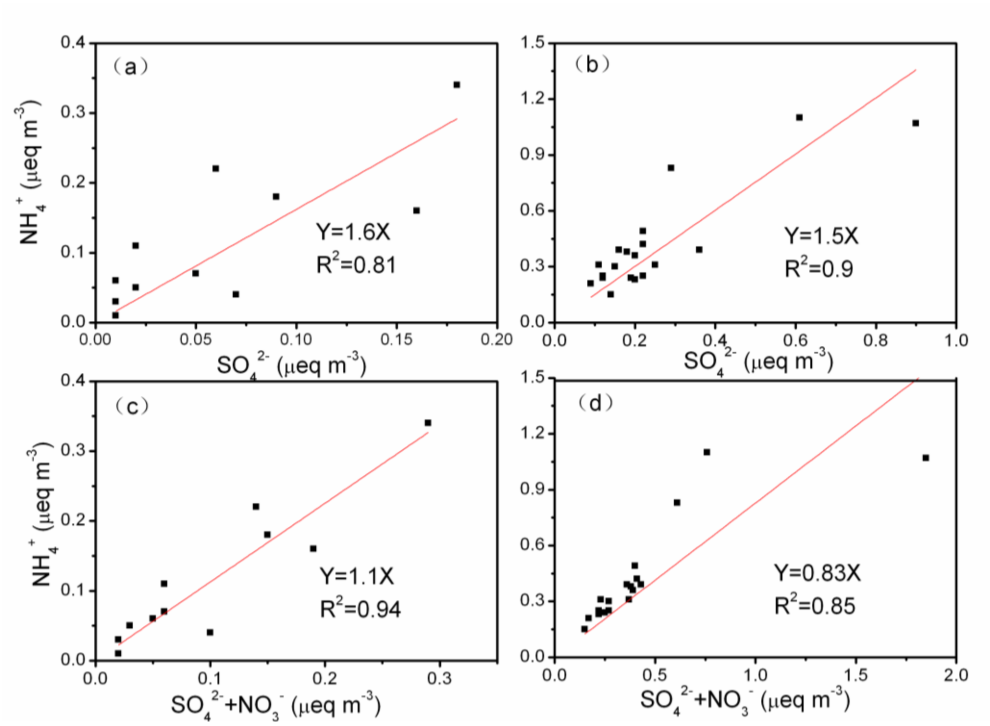


Fig. S6 Correlations between certain cations and anions in fine particles on both non-haze and haze days: (a) NH_4^+ and SO_4^{2-} on non-haze days; (b) NH_4^+ and SO_4^{2-} on haze days;(c) $NH_4^+$to [NO_3^- + SO_4^{2-}] on non-haze days; (d) $NH_4^+$to [NO_3^- + SO_4^{2-}] on haze days.

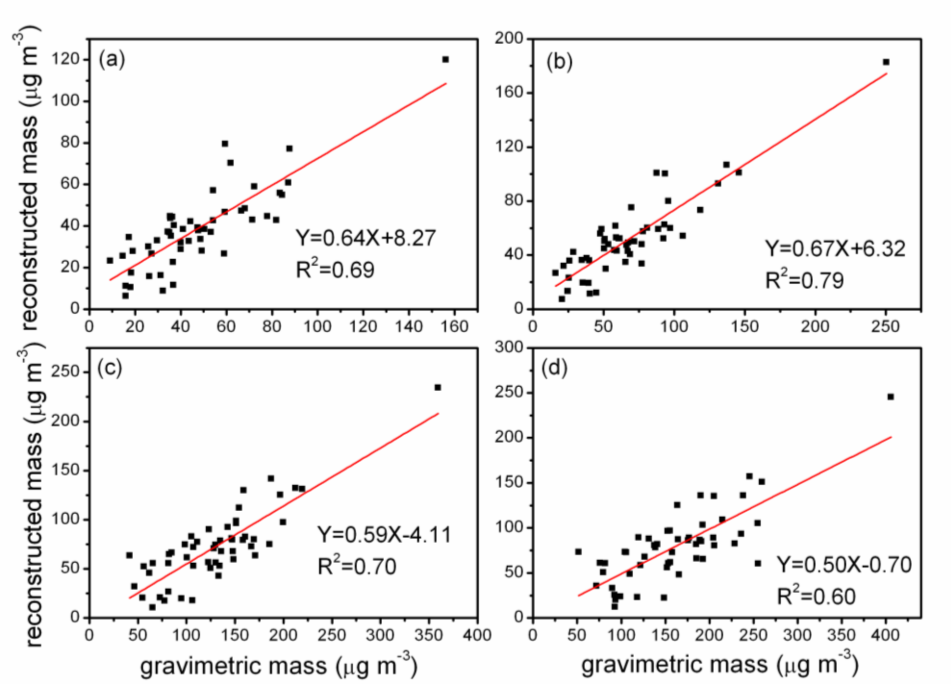


Fig.S7 The relationship between reconstructed PM mass concentrations and the gravimetric PM mass concentrations: (a) $PM_{1.1}$; (b) $PM_{2.1}$; (c) PM_9; (d) TSP.

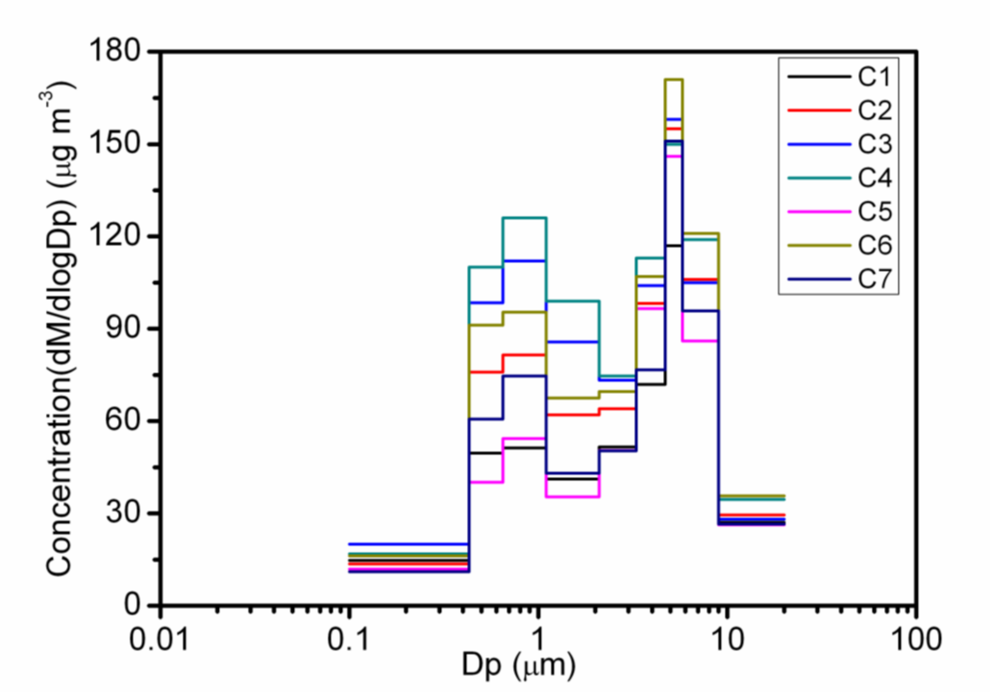


Fig. S8 Mass concentration size distributions within each trajectory cluster.

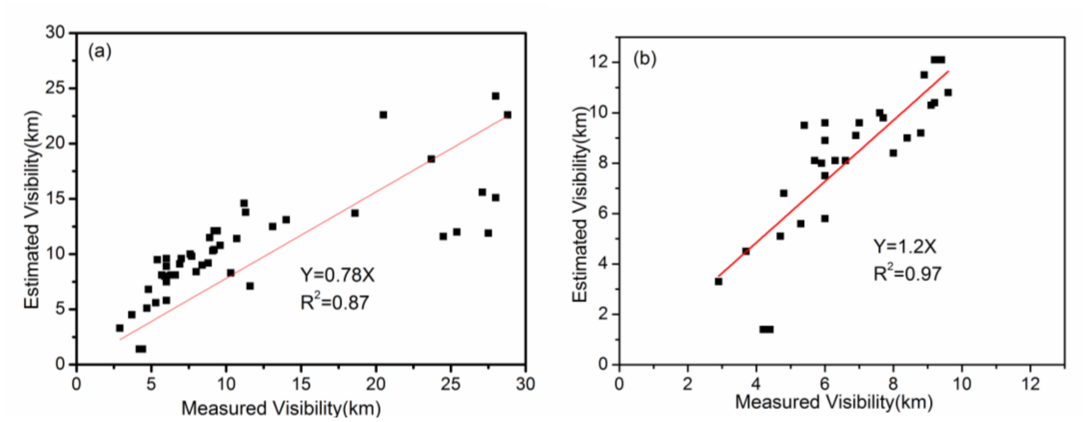


Fig. S9 Correction between estimated visibility and measured visibility (a) all the datasets in 2012-2013; (b) datasets with visibility lower than 10 km in 2012-2013.

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