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Increased inorganic aerosol fraction contributes to air pollution and haze in China

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1.Method to calculate volume fraction of ammonium sulfate and ammonium nitrate

we applied a commonly accepted ion pairing scheme of calculating the neutral aerosol from the molar number of all ions simplified by Gysel et al. (2007). In this scheme, by setting the fraction of nitric acid to zero, the molar fraction of ammonium nitrate is equal to the molar fraction of the measured nitrate ions. The rest of ammonium ions are assigned to ammonium sulfate, ammonium bisulfate and sulfuric acid according to ammonium to sulfate ratio. The aerosol in our study contained no sulfuric acid after the ion pairing scheme. To convert the mole fraction of these compounds to corresponding volume fractions, bulk densities for these chemical species were listed in Table 1, and the hygroscopic parameters of individual species were used based on literature values. Finally, a ZSR (Zdanovskii Stokes Robinson) mixing role was applied for the mixture, assuming internal mixing.

Table1 Hygroscopic growth factors kappa (κ) and density (ρ) for pure substance used in the Köhler theory

Substance	**κ (at α_ω =0.85)**	**ρ[kg/m^3]**
NH_4NO_3	0.68[a]	1720[b]
$(NH_4)_2SO_4$	0.53[a]	1769[b]
NH_4HSO_4	0.56[a]	1780[b]
H_2SO_4	0.97[c]	1830[b]
Organic	0.1[b]	1400

a Petters and and Kreidenweis (2007)

b Duplissy et al. (2011)

c Biskos et al. (2009)

d Hallquist et al. (2009)

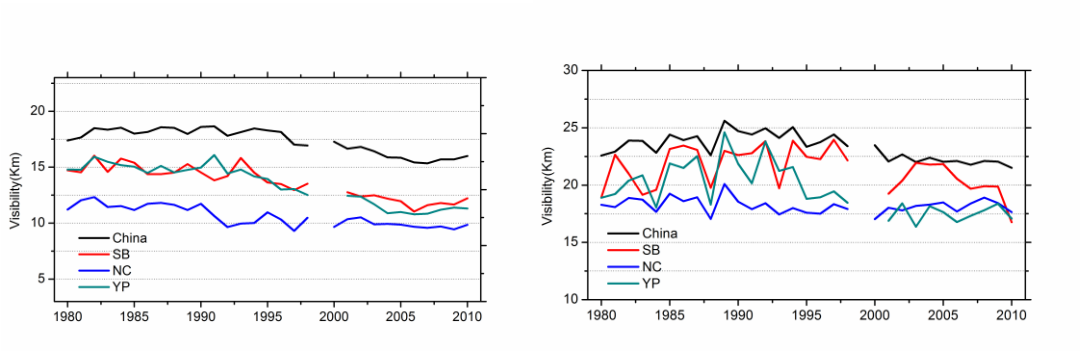
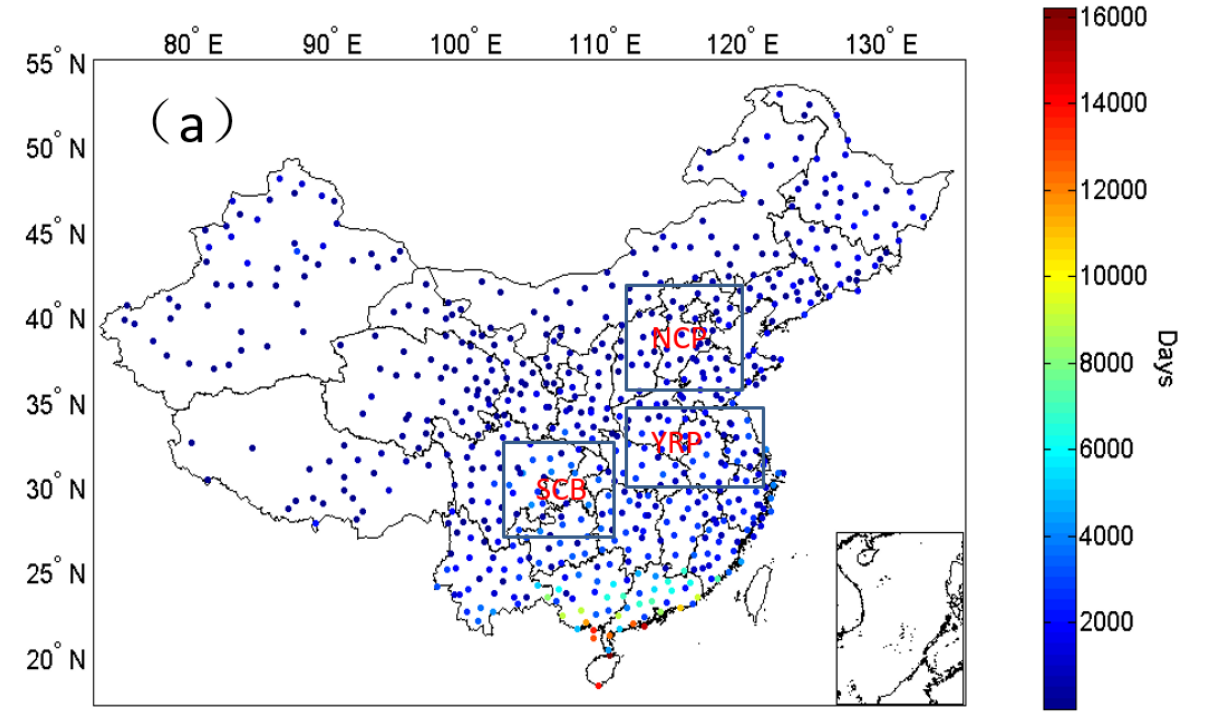


Figure s1. Long-term trends of visibility trends over Sichuan Basin (SCB), Northern Plain (NCP), Yangtze Plain (YRP) and China in low RH conditions and high RH conditions.


(a)
80° E
90° E
100° E
110° E
120° E
130° E
55° N
50° N
45° N
40° N
35° N
30° N
25° N
20° N
NCP
YRP
SCB
16000
14000
12000
10000
8000
6000
4000
2000
Days

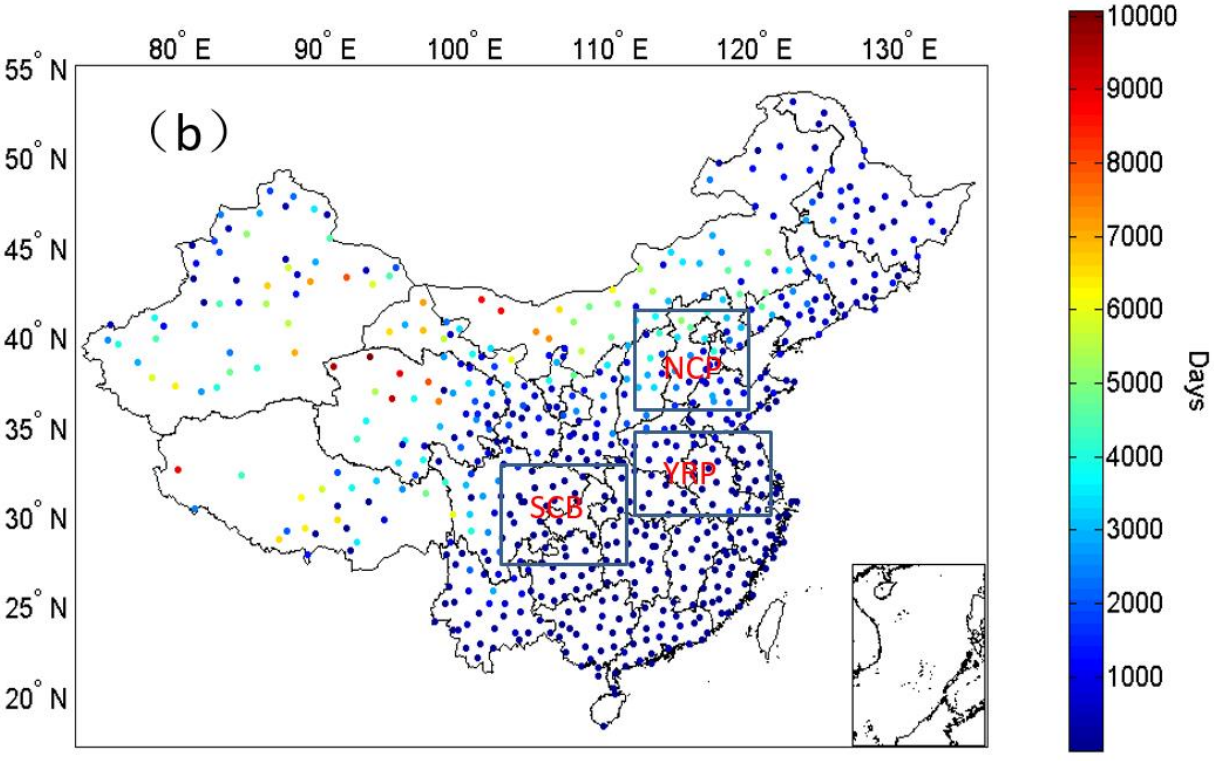

(b)
80° E
90° E
100° E
110° E
120° E
130° E
55° N
50° N
45° N
40° N
35° N
30° N
25° N
20° N
NCP
YRP
SCB
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
Days

Figure s2. The distribution of (a) high relative humidity days (80%~90%) and (b) low relative humdity days (<40%) from 1980-2010.

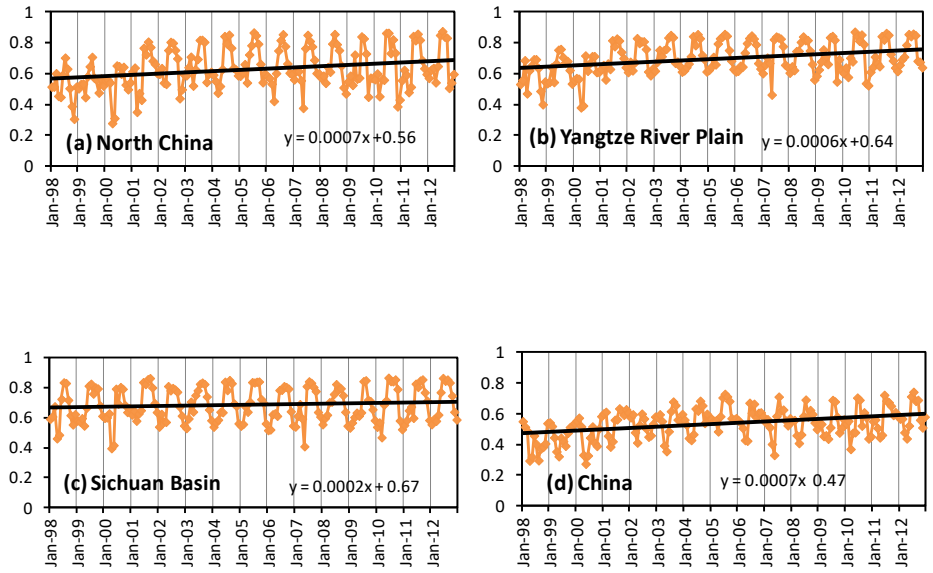


Figure s3. Trends of modeled inorganic aerosol fraction over China from the year 1998 to 2012

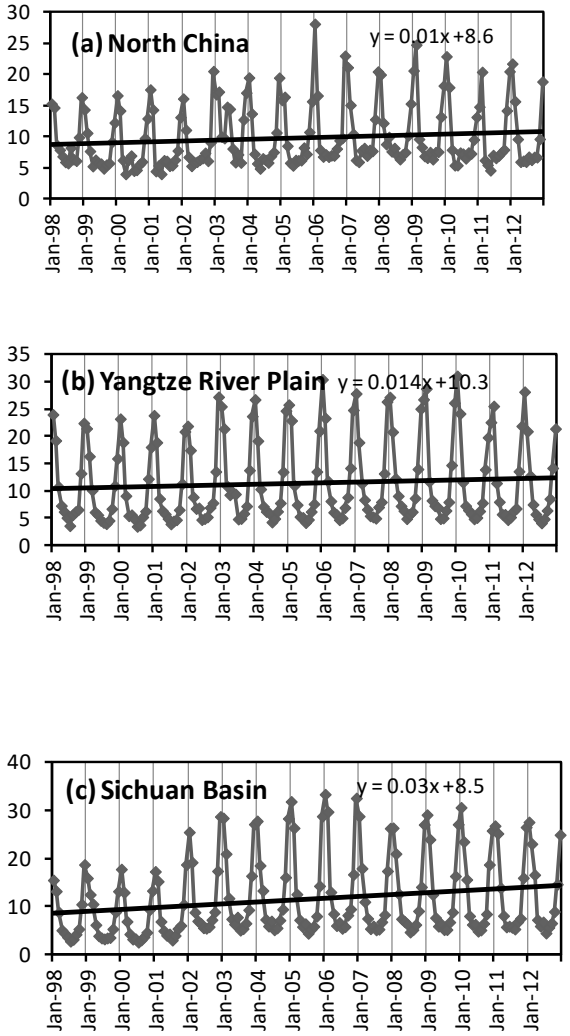


Figure s4. Trends of modeled carbonaceous aerosol concentrations over China from the year 1998 to 2012 (μg m^{-3})

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