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Supporting Information for:

An unexpected catalyst dominates formation and radiative forcing of regional haze

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Tables S1 and S2 (Tables S1 and S2 are referenced in the main manuscript)
Figs. S1 to S7 (Figs. S1 through S7 are referenced in the main manuscript)

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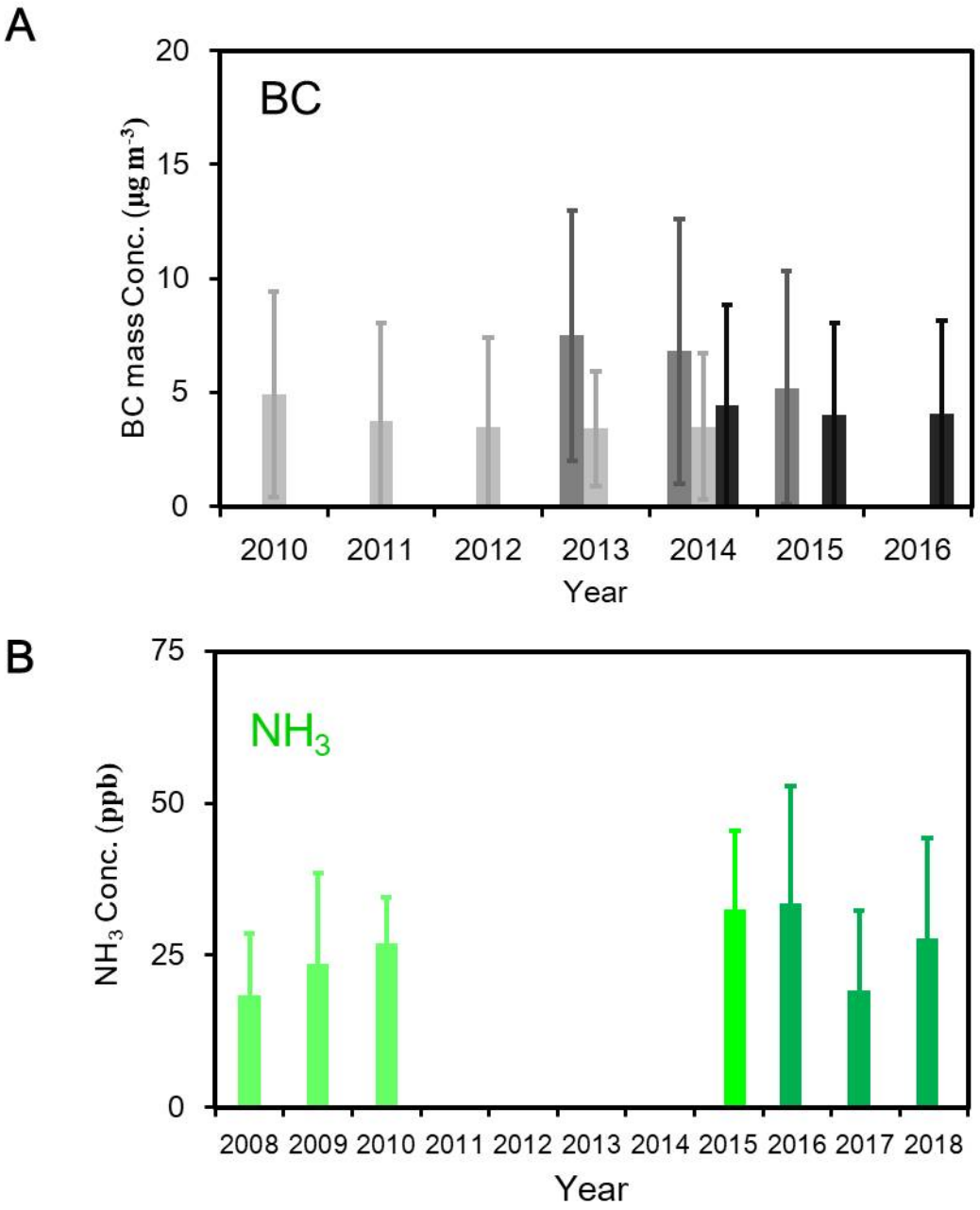

Fig. S1. Long-term trends of BC and NH_3 in Beijing. (**A** and **B**) Measurements of BC (A) and NH_3 (B). In (A), the BC mass concentrations marked by light-grey, grey, and black columns are taken from Liu et al. (39), Emilenko et al. (40), and measurements at PKU (16), respectively. In (B), the NH_3 concentrations marked by light-green, green, and dark-green columns are taken from Meng et al. (41), measurements at CMA, and Wang et al. (13), respectively.

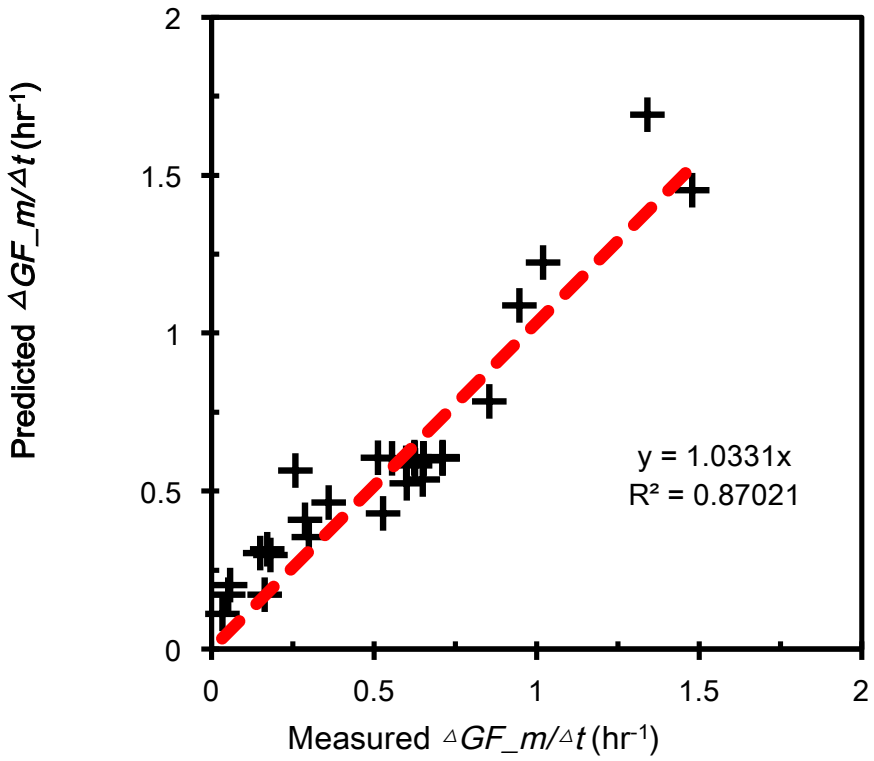

Fig. S2. Parameterization of the BC-catalyzed sulfate production rate. Comparison between the calculated and measured growth rates from the experimental results shown in Fig. 2, with a correlation coefficient of 0.87. The calculated value is based on the parameterization from laboratory experiments (see Methods).

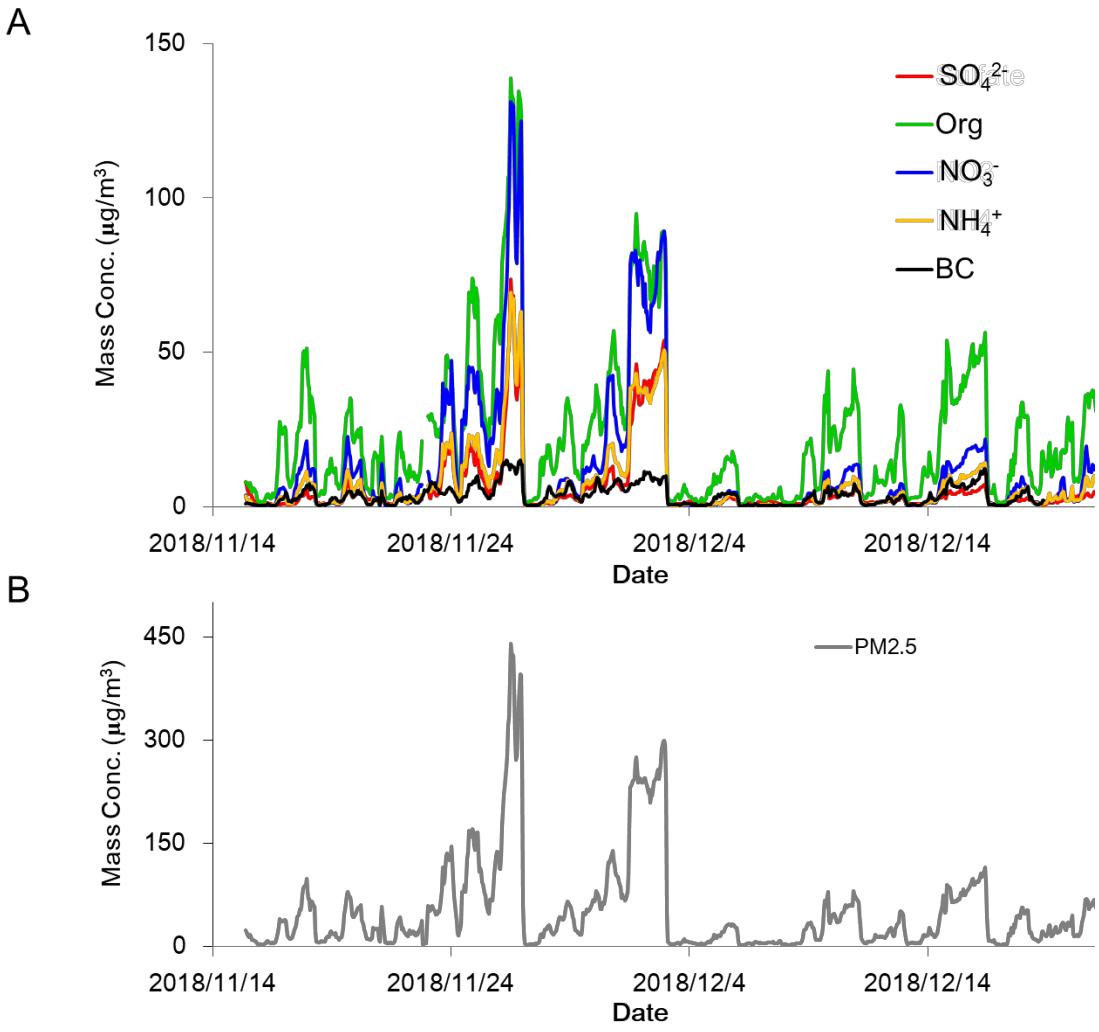

Fig. S3. $PM_{2.5}$ and chemical composition during 2018 winter Beijing. (**A**) Measurements of various the chemical compositions in $PM_{2.5}$ by AMS from November 15 to December 25, 2018. (B) $PM_{2.5}$ during the same period. The average $PM_{2.5}$ mass concentration is 53 µg m^{-3}, and the $PM_{2.5}$ ranges from less than 10 to 440 µg m^{-3} during this period. There were two heavy haze episodes on November 27-30 and December 1-3, with the maximum $PM_{2.5}$ mass concentration of 150 and 300 µg m^{-3}, respectively.

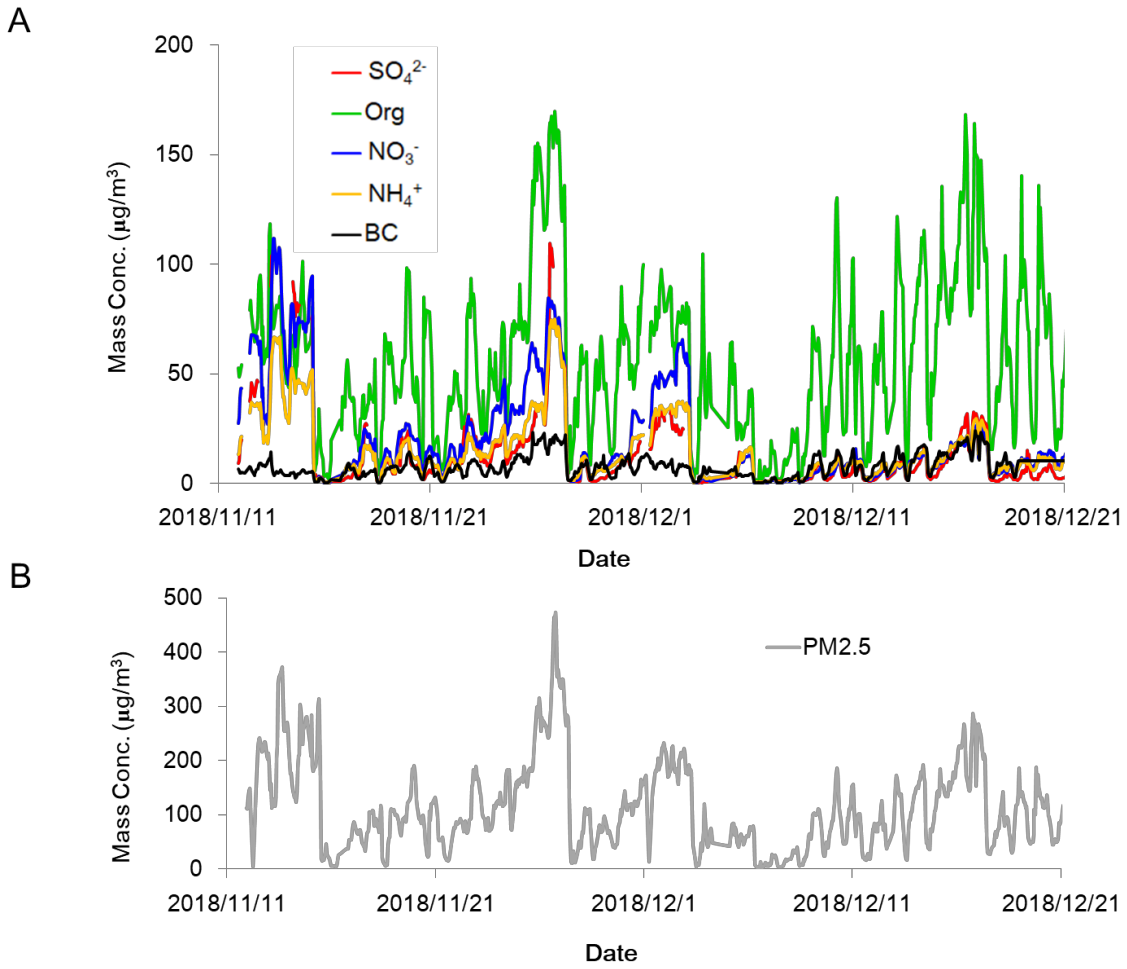

Fig. S4. $PM_{2.5}$ and chemical composition in 2018 winter Gucheng. (**A**) Measurements of the various chemical compositions in $PM_{2.5}$ by AMS from November 11 to December 21, 2018. (B) $PM_{2.5}$ during the same period. The average $PM_{2.5}$ mass concentration is 112 μg m^{-3}, and the $PM_{2.5}$ ranges from less than 10 to 490 μg m^{-3} during the period. There were two heavy haze episodes on November 24-26 and November 28 - December 1, with the maximum $PM_{2.5}$ mass concentration of 224 and 473 μg m^{-3}, respectively.

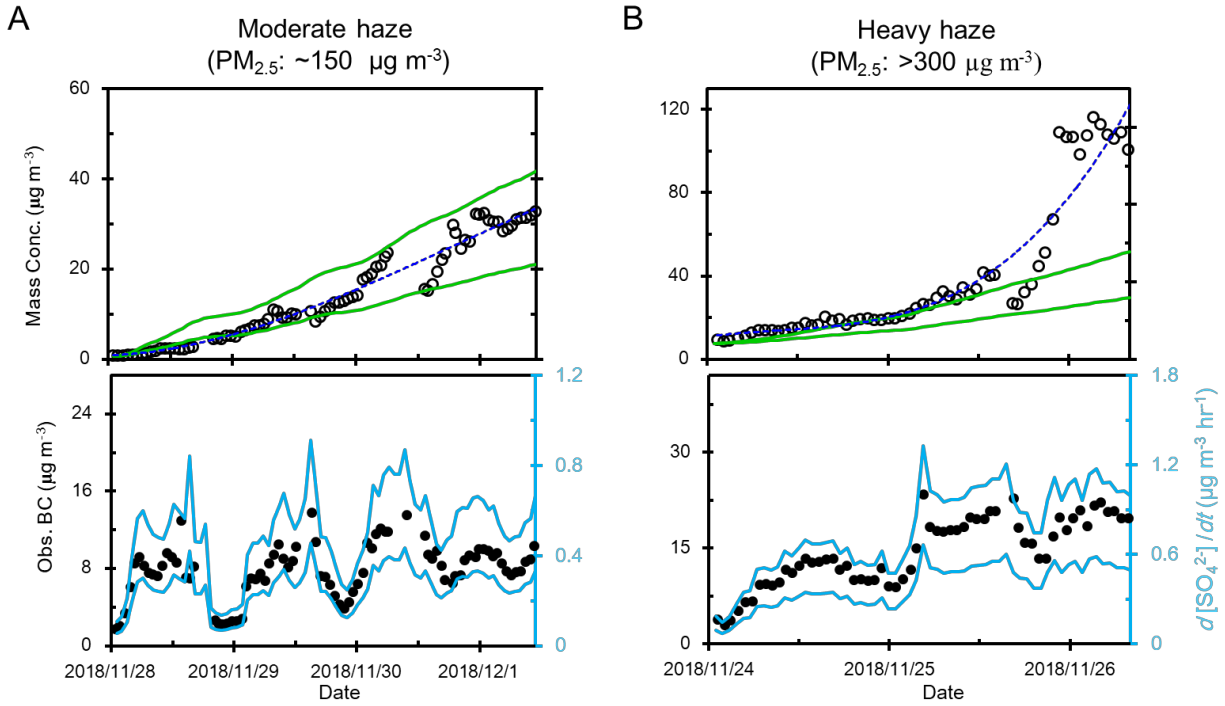

Fig. S5. Quantifying BC-catalyzed sulfate formation in Gucheng. (**A** and **B**) Calculated (green lines) and measured (circles and dashed line) sulfate mass concentrations during a moderate day and a heavy haze day, respectively. (**C** and **D**) BC mass concentration (left axis) and sulfate formation rate ($d[SO_4^{2-}]/dt$, right axis) during a moderate day and a heavy haze day, respectively. The range of the BC estimation (green and blue lines) are derived by assuming that 40% and 20% of the measured BC mass concentration are freshly emitted (34). Since there were no measurements of gaseous NH_3 during the observation period in Gucheng, we assumed a NH_3 concentration of 20 and 30 ppb during moderate and heavy haze periods, respectively, based on previous data in China (4,13).

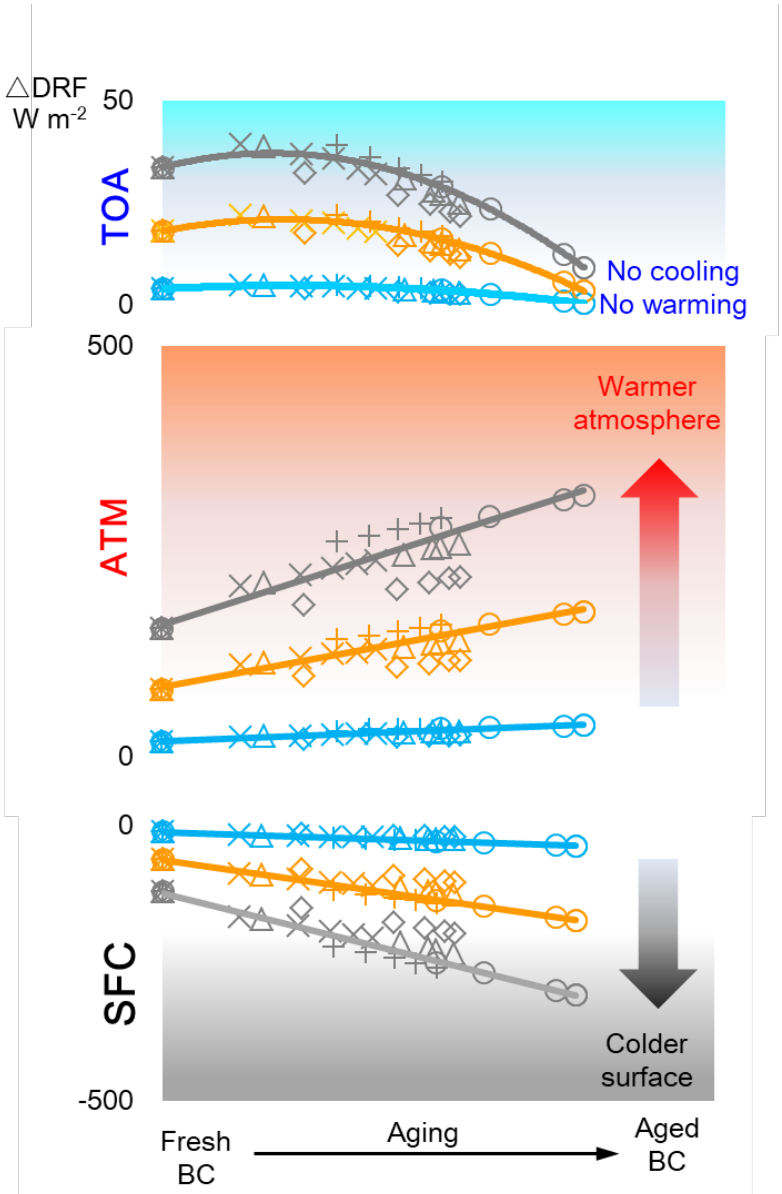

Fig. S6. ΔDRF dependence on BC aging at TOA, ATM and SFC on clean/light haze (blue lines), moderate haze (orange lines), and heavy haze (grey lines) days. The ambient average BC concentrations are assumed to be 2, 5, and 10 μg m^{-3}, on clean/light haze, moderate haze, and heavy haze days, respectively. A core−shell Mie model is adopted (35). The calculation is made at 532 nm with a unit of W m^{-2} μm^{-1}. The initial fresh BC diameter is 150 nm.

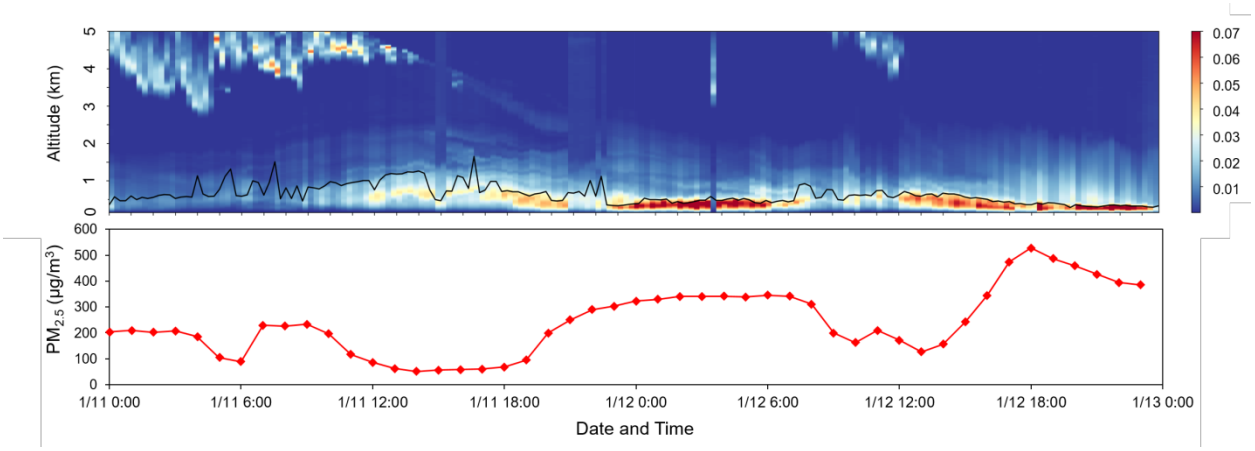

Fig. S7. PBL and $PM_{2.5}$ in Beijing. (**A**) The PBL height (solid line) derived from Lidar measurements during a haze event (January 11-12, 2019). (**B**) Mass concentration of $PM_{2.5}$ during the same period.

Table S1. Measured Gfm (within 2 hrs) and derived d(Gfm-1)/dt (in hr^{-1}) at variable concentrations of SO_2, NO_2, and NH_3 as well as RH.

Variables	Conc. (ppb)	Gfm (within 2 hrs)	d(Gfm-1)/dt (hr^{-1})
SO_2[a]	7.5	2.18	0.62
	37.5	2.07	0.60
	90*	1.40	0.20
	90*	1.80	0.40
	180*	1.40	0.20
	300	2.09	0.55
	375	2.23	0.65
	750	2.38	0.71
	750	2.26	0.62
NO_2[b]	37.5	0.95	0.02
	37.5	1.26	0.16
	180*	1.40	0.20
	375	2.23	0.65
	375	2.38	0.71
	375	2.26	0.62
	750	2.93	0.95
NH_3[c]	100	1.59	0.30
	180*	1.40	0.20
	300	1.40	0.20
	500	2.23	0.65
	1000	2.64	0.85
Relative humidity (RH)[d]	12%	1.00	0.00
	21%	1.19	0.10
	30%	1.36	0.18
	41%	1.59	0.29
	43%	2.02	0.51
	56%	2.34	0.67
	68%	2.23	0.62
	70%	2.26	0.63
	70%	2.38	0.69
	81%	2.20	0.60
	95%	2.26	0.63

[a]With NO_2, NH_3 and RH of 375 ppb, 500 ppb, and 70% for each experiment ;

[b]With SO_2, NH_3 and RH of 375 ppb, 500 ppb and 70% for each experiment;

[c]With SO_2, NO_2, and RH of 375 ppb, 375 ppb and 70% for each experiment;

*The data were not included in deriving the parameterization, but were used for validating the performance of the fitted parameterization. The experiments were performed in the SO_2, NO_2, NH_3 concentrations of 180-500 ppb and RH of 70%-90%.

Table S2. Gaseous and PM pollutants and meteorological parameters in Beijing and Gucheng.

	Chemical components in $PM_{2.5}$ and $PM_{2.5}$ ($\mu g\ m^{-3}$)							Meteorological parameters			Trace gases (ppb)				
	SO_4^{2-}	Org	NO_3^-	NH_4^+	Cl	BC	$PM_{2.5}$	T (^oC)	RH (%)	Wind ($m\ s^{-1}$)	O_3	CO	SO_2	NO_2	NH_3
Beijing															
Moderate haze	4.7±3.3	24.9±12.0	11.8±11.2	7.1±5.2	1.1±0.8	4.8±2.0	54.5±32.5	4.4±2.2	34.9±11.5	2.1±0.7	3.4±3.3	1.1±0.3	5.9±3.5	43.9±11.3	19.6±5.5
Heavy haze	32.6±15.2	68.3±20.8	61.9±23.6	33.4±12.6	3.6±1.9	8.2±1.7	208.1±72.8	5.2±1.5	72.4±16.1	1.9±0.8	5.0±7.0	1.7±0.6	3.7±2.0	60.4±10.1	36.1±8.7
Gucheng															
Moderate haze	14.1±10.8	61.3±22.3	24.2±16.6	17.7±10.9	5.1±1.9	7.8±2.8	128.7±60.3	2.1±3.6	77.5±18.8	0.3±0.5	4.2±4.8	1.6±0.5	8.0±1.7	34.9±7.6	NA
Heavy haze	40.2±34.8	108.2±43.4	52.2±18.6	37.1±19.5	11.6±5.9	14.5±5.3	264.0±119.9	2.4±4.3	81.3±20.1	0.6±0.8	5.2±6.6	2.2±0.7	10.3±5.8	49.7±12.6	NA